THE PROCEEDINGS OF
THE PHYSICAL SOCIETY

Section A

VOL. 65, PART 12 1 December 1952 No. 396A

The Vibrational Overlap Integral and the Intensities in Band Systems of Diatomic Molecules*

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MS. received 3rd June 1952

ABSTRACT. For the evaluation of the so-called vibrational overlap integral in the theory of the relative intensities in a band system of a diatomic molecule various approximate methods have been proposed for obtaining the vibrational wave functions. The ' distorted wave ' method of Gaydon, Pearse and Pillow is here discussed. For the case of Morse oscillators an approximate method is suggested that enables the integrals to be evaluated semi-analytically. The method is applied to the calculation of the relative intensities of the second negative bands of O_2^+ studied recently experimentally by Feast. A procedure for applying the WKB method to find the vibrational wave functions of an anharmonic oscillator is also given. A general exact relation involving the vibrational wave functions of two electronic states and their potentials is given which may be used as a check for the accuracy of the approximate wave functions obtained by various methods.

§ 1. INTRODUCTION

IN many problems involving the estimation of the temperature of the emitting gas (stellar atmosphere, aurora, night sky) from the relative intensities of the bands of a molecule it is necessary to have a theoretical knowledge of the relative transition probabilities for the various bands in a band system. The theory of these relative intensities is well known. To the approximation in which the dependence of the electronic moment of the molecule on the nuclear motion is neglected, the relative intensities of the bands of a system contain as a factor the square of the following so-called overlap integral,

$$I(n'v'\,;\ n'',v'') = \int_0^\infty \psi_{n'v'}(r)\psi_{n''v''}(r)\,dr, \qquad \ldots\ldots(1)$$

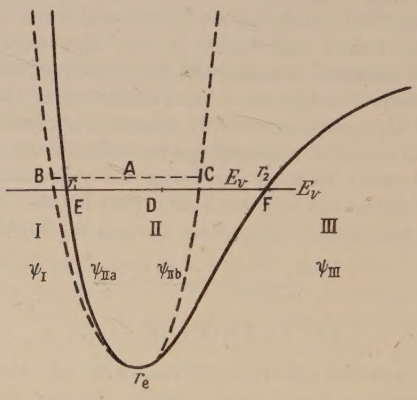
where $\psi_{n'v'}(r)$ is the vibrational wave function of the electronic state n' and the vibrational state v', r being the nuclear separation, etc. While one must bear in mind the approximate nature of the theory in which the relative intensities contain as a factor the square of $I(n',v'\,;\ n'',v'')$, it is nevertheless useful to have a fairly accurate but not too laborious method of evaluating the integral (1).

* Contribution from the National Research Council of Canada.

In those cases where laboratory data on the relative intensities of the bands in a system are available, a comparison with fairly reliable values of the integral (1) will at least indicate the extent to which the approximation (neglect of dependence of the electric moment on the nuclear separation) is valid, whereas no definite conclusion can be drawn from a comparison with admittedly rough estimates of the integral (1). Various methods for the evaluation of (1) have been proposed by various authors. The purpose of the present note is to discuss some of these methods and to suggest the procedures of two approximate methods that are free from the objections to the prevalent one used by many authors.

§ 2. THE METHOD OF DISTORTED WAVES

Let us first consider the procedure of Pillow* (1949, 1950, 1951) for obtaining an approximate wave function of an anharmonic oscillator from the corresponding wave function of a harmonic oscillator. The procedure consists in (i) locating the 'centre' D of the anharmonic potential (for a given state v) defined by that node or that maximum or minimum of the wave function $\psi_v(r)$ which corresponds to the centre of the harmonic oscillator, (ii) distorting the wave function of the harmonic oscillator by displacing its centre to D, compressing the abscissa to the left of the centre by the ratio DE/AB and



stretching the abscissa to the right by the ratio DF/AC, as indicated in the figure. The advantage of this procedure is obviously the availability of the wave functions of the harmonic oscillator. For transitions involving only the low vibrational states of both electronic states this procedure may be expected to be fairly satisfactory.

In principle, however, this procedure has the following disadvantages, which are the more serious the higher the vibrational states involved:

(i) The wave functions so obtained for the various vibrational states of the anharmonic oscillator are obviously no longer orthogonal, that is

$$\int_0^\infty \psi_{nv'}(r)\psi_{nv''}(r)\,dr \neq 0.$$

This lack of orthogonality will introduce some error in the relative values of $I(n', v'; n'', v'')$.

* The method described by Pillow is a refinement of one first suggested by Gaydon and Pearse (1939). As Pillow's procedure has removed some of the most serious objections that can be raised against the original method, we shall confine our discussion here to the procedure described by Pillow (1951).

(ii) Consider next the amplitude of the vibrational wave function. For any state v the function $\psi_v(r)$ should have a greater amplitude near the outer turning point r_2 (F in the figure) than near the inner turning point r_1 (E in the figure). This is a consequence of the general shape of the potential curve which is steep at r_1 and flat at r_2. Now the wave functions obtained by the distortion procedure described above have the same amplitude near both turning points. It is seen that the error introduced on this account increases as the vibrational quantum number increases.

(iii) Consider finally the 'centre' D of the actual potential $V(r)$. As the integral (1) depends quite critically on the overlap or phase relationship of the two wave functions $\psi_{n'v'}(r)$ and $\psi_{n''v''}(r)$, it is important that the phases of the wave functions of the various states be accurately determined. The recent work of Pillow achieves an important refinement of the earlier procedure of Gaydon and Pearse (1939) in allowing for the displacement of the distorted wave functions to the centre D of the actual potential.* In principle, the centre D for any state v can be located for the Morse oscillator, but it is seen from the wave function (5) that for the state v one has to obtain the roots of an equation of degree v. If one plots the wave functions (5) to determine the centre D, one already has the wave functions $\psi_v(r)$, and there will be no need for the 'distorted' wave functions. The plotting of the wave functions (5), however, for a large number of states v is the lengthy calculation that we wish to replace by approximate methods.

§ 3. A GENERAL RELATION

We shall now first give a general relation involving the vibrational wave functions $\psi_{n'v'}(r)$, $\psi_{n''v''}(r)$ of two electronic states with potentials $V_{n'}(r)$ and $V_{n''}(r)$. The Schrödinger equations are

$$\frac{d^2\psi^*_{n'v'}}{dr^2} + \frac{8\pi^2\mu}{h^2}[E_{n'v'} - V_{n'}(r)]\psi^*_{n'v'} = 0,$$

$$\frac{d^2\psi_{n''v''}}{dr^2} + \frac{8\pi^2\mu}{h^2}[E_{n''v''} - V_{n''}(r)]\psi_{n''v''} = 0. \qquad \ldots\ldots(2)$$

On multiplying the first equation by $\psi_{n''v''}(r)$, the second by $\psi^*_{n'v'}(r)$, subtracting one resulting equation from the other and integrating over the coordinate r, one obtains the relation

$$(E_{n'v'} - E_{n''v''}) \int_0^\infty \psi^*_{n'v'}\psi_{n''v''}\, dr = \int_0^\infty \psi^*_{n'v'}[V_{n'} - V_{n''}]\psi_{n''v''}\, dr, \quad \ldots\ldots(3)$$

which is exact. While eqn. (3) cannot be used directly for the evaluation of the integral $I(n', v'; n'', v'')$, it is a useful relation that serves as a check for the accuracy of the approximate wave functions $\psi_{n'v'}$, $\psi_{n''v''}$ that have been obtained by various approximate methods.

* It is stated by Pillow (1951) that the distance a(OD) (see her fig. 3) is given, up to the first power in x_e, by the relation $a(\mathrm{OD}) = (v+1)x_e$, so that the locus of D for the various states v it nearly a straight line. Calculation with the wave function (5) for the Morse oscillator showss however, that the distance a(OD) increases by more than x_e for each unit increase in v for large v, as shown below :

v	0	1	2	3	9
$a(\mathrm{OD})/x_e$	1	2	$\sim 3+\frac{2}{3}$	$\sim 4+\frac{2}{3}$	~ 13

Thus the locus of D bends towards larger r as v increases. This bending is very great for large v.

§ 4. MORSE OSCILLATOR

For many molecules it is convenient and sufficient to represent the potential of a given electronic state by a Morse function

$$V(r) = \mathrm{D}[1 - \exp\{-a(r-r_e)\}]^2. \qquad \ldots\ldots(4)$$

It is well known that for this potential the wave functions are given by

$$\psi_v(z) = N_v e^{-z/2} z^{\beta/2} L^{\beta}_{v+\beta}(z), \qquad \ldots\ldots(5)$$

where $x_e z = \exp\{-a(r-r_e)\}$, $\beta = x_e^{-1} - 2v - 1$, $x_e = \hbar\omega\mathbf{c}/4D$,

$$L^{\beta}_{v+\beta}(z) = \sum_{k=0}^{v} \frac{(-1)^k \Gamma(\beta+v+1)}{k!\,(v-k)!\,\Gamma(\beta+k+1)} z^k, \quad N_v^2 = a\beta v!/\Gamma(\beta+v+1).$$

Let us denote the various quantities in (5) for the two states n', v' and n'', v'' by a single and a double prime respectively, and let us choose z' as the variable in the integral (1) and omit this prime from z' for brevity. Then

$$dr = -dz'/a'z' = -dz/a'z, \quad z'' = \xi z^{\gamma},$$

where

$$\xi = \frac{1}{x_e''}(x'_e)^{\gamma} \exp\{a''(r_e'' - r_e')\}, \quad \gamma = a''/a'. \qquad \ldots\ldots(6)$$

The integral (1) can then be shown to be

$$I(n', v'; n'', v'')$$

$$= (-1)^{v'+v''} \frac{N_{v'} N_{v''}}{a'} \xi^{1/2 x_e'' - 1} \sum_{\lambda=0}^{v'} \sum_{\mu=0}^{v''} (-1)^{\lambda+\mu} B(v', \lambda) B(v'', \mu) I(\lambda, \mu) \qquad \ldots\ldots(7)$$

where

$$B(v', \lambda) = \frac{(\beta'+v')}{\lambda!\,(v'-\lambda)!}, \quad B(v'', \mu) = \frac{(\beta''+v'')_{\mu}}{\mu!\,(v''-\mu)!\,\xi^{\mu}} \qquad \ldots\ldots(8)$$

$$(\beta+v)_{\lambda} \equiv (\beta+v)(\beta+v-1)\ldots(\beta+v-\lambda+1), \qquad (\beta+v)_0 \equiv 1,$$

and

$$I(\lambda, \mu) = \int_0^{\infty} \exp\{-\tfrac{1}{2}(z + \xi z^{\gamma})\} z^p \, dz, \qquad \ldots\ldots(9)$$

$$p = \frac{1}{2x_e'} + \frac{\gamma}{2x_e''} - 1 - \tfrac{1}{2}(1+\gamma) - \lambda - \gamma\mu. \qquad \ldots\ldots(10)$$

The integrals $I(\lambda, \mu)$ in (9) cannot be evaluated in analytic form in general. While a numerical or graphical evaluation is possible, the large number, $(v'+1)(v''+1)$, of such integrals involved in a transition v', v'' makes such a procedure impracticable. The following method is here suggested. The integrand in (9) has a rather sharp maximum at z_0 given by

$$z_0 + \xi\gamma z_0^{\gamma} = 2p \qquad \ldots\ldots(11)$$

and falls off rather symmetrically for $z < z_0$ and $z > z_0$. Nearly all the contribution to the integral (9) comes from the neighbourhood of z_0. Let us replace the integrand for $z > z_0$ by a gaussian function

$$G_1(z) = A \exp\{-\alpha_1^2 (z - z_0)^2\} \qquad \ldots\ldots(12)$$

with A equal to the maximum value of the integrand at z_0, namely,

$$A = z_0^{p} \exp\{-\tfrac{1}{2}(z_0 + \xi z_0^{\gamma})\}, \qquad \ldots\ldots(13)$$

and α_1^2 so chosen that at a point z_1 where the integrand falls to $\frac{1}{2}A$, $G_1(z_1) = \frac{1}{2}A$. This condition gives the relations

$$z_0(\epsilon_1 - 1) + \xi z_0^{\gamma}(\epsilon_1^{\gamma} - 1) - 2p \ln \epsilon_1 = 2 \ln 2, \qquad \ldots\ldots(14)$$

$$\alpha_1^2 = \ln 2 / z_0^2 (\epsilon_1 - 1)^2, \quad \epsilon_1 = z_1/z_0 > 1. \qquad \ldots\ldots(15)$$

Similarly, for the region $z<z_0$, we replace the integrand in (9) by $G_2(z)=A\exp\{-\alpha_2^2(z-z_0)^2\}$, with α_2^2 given by

$$z_0\left(1-\frac{1}{\epsilon_2}\right)+\xi z_0^{\gamma}(1-\epsilon_2^{-\gamma})-2p\ln\frac{1}{\epsilon_2}=-2\ln 2, \quad \ldots\ldots(14a)$$

$$\alpha_2^2=\ln 2/z_0^2\left(1-\frac{1}{\epsilon_2}\right)^2, \quad \epsilon_2=z_0/z_2>1. \quad \ldots\ldots(15a)$$

The integral (9), in which the integrand has been approximated by $G_2(z)$ for $z<z_0$ and by $G_1(z)$ for $z_0<z$, can be evaluated in a simple form, and by means of (13), (15), (15 *a*) one obtains

$$I(\lambda,\mu)=\tfrac{1}{2}\left(\frac{\pi}{\ln 2}\right)^{1/2}\left(\epsilon_1-\frac{1}{\epsilon_2}\right)z_0^{p+1}\exp\{-\tfrac{1}{2}(z_0+\xi z_0^{\gamma})\}. \quad \ldots\ldots(16)$$

The accuracy of this approximation has been tested for the case of the second negative bands of O_2^+, which we shall describe more fully below. It is found that the value given by (16) differs from the value of the integral (9) obtained by graphical integration by less than one per cent.

Table 1. Molecular Constants of the $A^2\Pi_u$, $X^2\Pi_g$ States of O_2^+ (Feast 1950) in (4) and (6)

State	r_e (Å)	x_e	a	D_{calc}(cm^{-1})
$'$ ($A^2\Pi_u$)	1·4089	0·01525	$2{\cdot}56\times10^8$	14740
$''$ ($X^2\Pi_g$)	1·1227	0·00882	$2{\cdot}85\times10^8$	53200

$\gamma=1{\cdot}11, \quad \xi=0{\cdot}50$

Table 2. Calculated Squares of $I(n', v'; n'', v'')$ according to (7) and (16)

Visual estimates of the relative intensities of the second negative bands of O_2^+ given by Feast (1950) are given in parentheses.

v' \ v''	0	1	2	3	4	5	6
0	$3{\cdot}0\times10^{-3}$	$5{\cdot}8\times10^{-2}$	0·45	2·21	8·02	16·2	87·5
						(8)	(7)
1	$2{\cdot}8\times10^{-2}$	0·39	2·49	9·8	28·2	26·5	
				(3)	(5)	(6)	
2	0·12	2·27	7·30	23·6	31·5		
		(1)	(2)	(7)	(8)		
3	0·35	3·45	14·2	48·4	70		
		(1)	(2)	(8)	(7)		
4	0·81	6·96	18·7	135			
		(3)	(6)	(7)			
5	1·63	12·0					
		(2)					
6	1·88						

To illustrate this method, calculation of the integral (1) has been carried out for the second negative bands of O_2^+ recently studied by Feast (1950).* The empirical data for the two electronic states are given in table 1. In table 2 are given the values of the square of the integral (1) calculated according to (7) and (16). The visual estimates of the relative intensities of the bands given by Feast are given in parentheses in table 2. It is seen that while no quantitative comparison can be made, the general intensity distribution observed agrees with the calculated.

* The author is indebted to Professor Pearse for suggesting the calculation for these bands of O_2^+.

It may be of interest to describe the actual procedure found most convenient in the calculation. From the empirical constants in table 1 and by means of (10) the values of p corresponding to various values of λ, μ ($\lambda=0, 1, \ldots, v'$; $\mu=0, 1, \ldots, v''$) are tabulated in a two-dimensional (λ,μ) table. By means of tables of logarithms* the values of z_0 given by (11) are found for these values of $p(\lambda,\mu)$. The values of $\ln z_0$ and $\frac{1}{2}(z_0+\xi z_0^{\gamma})$ are next tabulated in this (λ,μ) table. By means of (14) curves of ϵ_1 and $1/\epsilon_2$ as functions of p are calculated, and from them a curve of $\ln(\epsilon_1-1/\epsilon_2)$ as a function of p is obtained. This curve is practically linear. Finally, since

$$\ln I(\lambda,\mu)=\tfrac{1}{2}\ln(\pi/\ln 2)+\ln\left(\epsilon_1-\frac{1}{\epsilon_2}\right)-\tfrac{1}{2}(z_0+\xi z_0^{\gamma})+(p+1)\ln z_0-\ln 2 \quad \ldots(17)$$

the logarithms of $I(\lambda,\mu)$ are tabulated in the (λ,μ) table. A useful check against accidental numerical errors is to plot $\ln I(\lambda,\mu)$ for constant $\lambda+\mu$ against p. These should be smooth curves.

Next the logarithms of $n!$, $(1/x_e'-1)$, $(1/x_e'-2)$, etc., are tabulated, from which one forms the arrays of

$$\left.\begin{aligned}\ln B(v',\lambda)&=\ln(\beta'+v')_{\lambda}-\ln\lambda!-\ln(v'-\lambda)!\\ \ln B(v'',\mu)&=\ln(\beta''-v'')_{\mu}-\ln\mu!-\ln(v''-\mu)!-\mu\ln\xi\end{aligned}\right\} \quad \ldots\ldots(18)$$

for all v', v'', and $\lambda=0, 1, \ldots, v'$, $\mu=0, 1, \ldots, v''$. From the (λ,μ) table and the tables (18), the logarithm of each term in the sum (7) is calculated.

§ 5. WKB METHOD

The method described in the last section depends on the use of the Morse potential. In some cases, such as H_2, it is not possible to represent, by means of the Morse potential, the observed vibrational levels up to high vibrational quantum numbers, together with the fundamental frequency ω, the anharmonicity factor x_e and the dissociation energy D. In such cases a numerically constructed potential or a function containing more adjustable parameters becomes necessary. To obtain fairly good vibrational wave functions in such cases the Wentzel–Kramers–Brillouin method* seems the most practical one. According to this method the vibrational levels of an oscillator with the potential $V(r)$ are given by the equation

$$\int_{r_1}^{r_2} P(r)\,dr=(v+\tfrac{1}{2})\pi, \quad \ldots\ldots(19)$$

where

$$P(r)=\frac{2\pi}{h}(2\mu)^{1/2}(|E_v-V(r)|)^{1/2}. \quad \ldots\ldots(20)$$

The wave function for the state v is given for the various regions indicated in the figure by the following expressions:

Region I, $r<r_1$

$$\psi_{\mathrm{I}}(r)=N_v P^{-1/2}\exp\{-Q(r;r_1)\},$$

Region II, $r_1<r<r_2$

$$\psi_{\mathrm{IIa}}(r)=2N_v P^{-1/2}\cos\{Q(r_1;r)-\tfrac{1}{4}\pi\}=$$
$$\psi_{\mathrm{IIb}}(r)=(-1)^v 2N_v P^{-1/2}\cos\{Q(r;r_2)-\tfrac{1}{4}\pi\}, \quad \ldots\ldots(21)$$

* Such as the *Tables of Natural Logarithms*, Work Project Administration, New York. The values of the sum in (7) are usually small compared with the individual terms. Hence it is necessary to retain six figures after the decimal in all the logarithms.

* See, for example, Condon and Shortley (1935).

Region III, $r_2 < r$

$$\psi_{III}(r) = (-1)^v N_v P^{-1/2} \exp\{-Q(r_2; r)\},$$

where $$Q(r; r_1) = \int_r^{r_1} P(r)\, dr, \text{ etc.},$$

and N_v is the normalization constant. The smooth joining of the two expressions for the wave functions in the region II is guaranteed by the condition (19). The sign $(-1)^v$ in (21), (23) also follows from (19).

The functions in (21) are good except in the immediate neighbourhood of the turning points r_1 and r_2, at which they diverge. In the immediate neighbourhood of each turning point, however, the potential $V(r)$ can be replaced by a linear function of r, i.e.

$$\left.\begin{array}{lll} \text{near } r_1, & P^2(r) = C(r - r_1), & C = \dfrac{8\pi^2\mu}{h^2}\left|\left(\dfrac{\partial V}{\partial r}\right)_{r_1}\right|, \\ \text{near } r_2, & P^2(r) = D(r_2 - r), & D = \dfrac{8\pi^2\mu}{h^2}\left|\left(\dfrac{\partial V}{\partial r}\right)_{r_2}\right|. \end{array}\right\} \quad \ldots\ldots (22)$$

For such potentials the Schrödinger equation can be solved exactly and the solutions are Bessel functions of order $\frac{1}{3}$ and $-\frac{1}{3}$. These solutions must be joined on to the solutions (21), which are valid beyond the immediate neighbourhood of r_1 and r_2. This condition of smooth joining determines the multiplying constant in these functions and also the values of the wave function at the points r_1 and r_2. The wave function for the various regions is then given as follows :

Region I

$$\psi_I(r) \longleftrightarrow N_v \left[\frac{2Q(r; r_1)}{\pi P(r)}\right]^{1/2} K_{1/3}\{Q(r; r_1)\} \longleftrightarrow bN_v C^{-1/6} \quad \text{at } r = r_1.$$

Region II

$$bN_v C^{-1/6} \longleftrightarrow N_v \left[\frac{2\pi Q(r_1; r)}{3P(r)}\right]^{1/2} [J_{1/3}\{Q(r_1; r)\} + J_{-1/3}\{Q(r_1; r)\}] \longleftrightarrow \psi_{IIa}(r) \longleftrightarrow$$

$$\psi_{IIb}(r) \longleftrightarrow (-1)^v N_v \left[\frac{2\pi Q(r; r_2)}{3P(r)}\right]^{1/2} [J_{1/3}\{Q(r; r_2)\} + J_{-1/3}\{Q(r; r_2)\}] \longleftrightarrow (-1)^v bN_v D^{-1/6} \quad \text{at } r = r_2 \quad \ldots\ldots (23)$$

Region III

$$(-1)^v bN_v D^{-1/6} \longleftrightarrow (-1)^v N_v \left[\frac{2Q(r_2; r)}{\pi P(r)}\right]^{1/2} K_{1/3}\{Q(r_2; r)\} \longleftrightarrow \psi_{III}$$

where $$b = (\tfrac{1}{3}\pi)^{1/2}\pi^{1/3}/\sin\tfrac{1}{3}\pi\Gamma(\tfrac{2}{3}) = 1{\cdot}26.$$

The wave function given in (23) looks more complicated than it actually will be in the calculation. For a given potential $V(r)$ and any given vibrational state v one plots $P(r)$ in (20) and obtains the functions $Q(r; r_1)$, $Q(r_1; r)$, etc., at suitable intervals of r by means of a graphical integration (with a planimeter, for example). The functions ($\psi_I(r)$, $\psi_{II}(r)$, $\psi_{III}(r)$ and the Bessel functions are readily calculated since they are analytic functions that have been tabulated. In the actual calculation it is sometimes even possible to join the wave function $\psi_I(r)$ and $\psi_{IIa}(r)$ smoothly over the singularity $r = r_1$ by making use of the finite

value $bN_v C^{-1/6}$ at r_1 without using the solutions involving the Bessel functions at all. Similarly for the point r_2. This procedure can be carried out to a good approximation after one has some experience with the behaviour of these various functions from a typical calculation. The normalization factor N_v is finally determined by another graphical integration.

The wave functions obtained by this WKB method can be expected to be better than those obtained by the 'distortion' method with regard to the phase relationship for the functions of various vibrational states and to the amplitude near the two turning points. Application of the procedure to certain bands of C_2 and CN of astronomical interest has been made by Wyller* (private communication). The result obtained seems better than that obtained with the use of the distortion method.

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On Jastrow's Interaction and the Elastic Scattering of High Energy Protons by Deuterons

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Communicated by H. S. W. Massey; *MS. received 9th July* 1952

ABSTRACT. An approximate method has been used to calculate the elastic total and differential cross sections for p–d scattering, at incident proton energies of 150 and 240 MEV. A nuclear interaction has been assumed of a type suggested by Jastrow, containing an infinite repulsive core in singlet two-body states. Of the various phenomenological interactions, this agrees about the best with the high energy n–p and p–p scattering data. The calculated angular distribution near 90°, in the centre-of-mass system, is found to be of the right order of magnitude, but agreement with experiment in the forward and backward directions is poor. An explanation of this result is suggested.

§ 1. INTRODUCTION

SEVERAL calculations of the high energy n–d scattering cross section have been reported in recent years (Wu and Ashkin 1948, Chew 1948, 1951, Bethe and Gluckstein 1951, Bransden 1951, to be referred to as I). Little agreement with experiment has been obtained, mainly for the reason that no direct evidence as to the nature of the neutron–neutron interaction is available. The potential functions representing this interaction, in these calculations, are therefore probably in error (indeed they are certainly in error if it is assumed, as seems likely, that the neutron–neutron and proton–proton interactions are identical). Recent experimental determinations of the elastic proton–deuteron

* Results obtained by Mr. Wyller were reported in a meeting of the American Astronomical Society, Victoria, B.C., in June 1952.

scattering cross section at 90, 150 and 240 MeV (Stern and Bloom 1951, Pickavance *et al.* 1951, Schamberger 1951) suggested that a calculation of this cross section would be of interest, using an interaction that reproduced the main features of proton–proton scattering.

The proton–proton collision cross section, although approximately independent of the angle of scattering, is too large to be due to pure S-wave scattering, unless internal changes take place in the colliding protons, as has been suggested by Breit (1951). Several interactions have been proposed that give results in approximate agreement with experiment, namely a singular tensor interaction (Christian and Noyes 1950), a singular spin–orbit coupling term (Case and Pais 1950) and an interaction containing an infinite repulsive core (Jastrow 1951). The common feature of these interactions is that at short distances they represent a short-range singular repulsive force. Perhaps the best agreement with experiment is given by Jastrow's interaction, which also may be able to produce saturation in heavy nuclei, and this interaction has therefore been used in the present calculation to find the proton–deuteron cross sections at incident proton energies of 150 and 240 MeV. Jastrow's interaction has the further advantage that it probably enables charge independence to be preserved. The calculations were not extended to cover the 95 MeV experimental data as the approximate method employed would certainly be in serious error at such energies.

Previous calculations of the elastic p–d scattering cross section have been reported by Chew (1951), using his impulse approximation. He found fair agreement with the 95 and 240 MeV experimental results in the forward direction. Instead of assuming an interaction as in the present work, the measured n–p and p–p cross sections were used.

Other calculations at 240 MeV were made by Auerbach and reported by Schamberger (1951). Auerbach used Born's approximation and assumed three alternative proton–proton interactions which, however, were inconsistent with the experimental p–p scattering data and, as expected, the calculated and experimental results did not agree.

§ 2. GENERAL THEORY

The interaction suggested by Jastrow is of the form

$$\begin{aligned} V^{\mathrm{S}}(r) &= \infty, \quad r<r_0 \\ &= \tfrac{1}{2}V_0^{\mathrm{S}}(1+M_{12})\exp\{-(r-r_0)/r_{\mathrm{s}}\}, \quad r>r_0 \qquad \ldots\ldots(1) \\ V^{\mathrm{T}}(r) &= V_0^{\mathrm{T}}\{a+(1-a)M_{12}+\{b+(1-b)M_{12}\}\gamma S_{12}\}\exp(-r/r_{\mathrm{t}}), \end{aligned}$$

where V^{S} and V^{T} are the two-body singlet and triplet interactions respectively, M_{12} is the space exchange operator, $S_{12}=3(\boldsymbol{\sigma}_1\cdot\mathbf{r})(\boldsymbol{\sigma}_2\cdot\mathbf{r})/r^2-\boldsymbol{\sigma}_1\cdot\boldsymbol{\sigma}_2$ is the tensor interaction operator and $r_0=0{\cdot}6\times10^{-13}$ cm, $r_{\mathrm{s}}=0{\cdot}4\times10^{-13}$ cm, $r_{\mathrm{t}}=0{\cdot}75\times10^{-13}$ cm, $a=0{\cdot}5$, $b=0{\cdot}3$, $\gamma=1{\cdot}84$, $V_0^{\mathrm{S}}=375$ MeV, $V_0^{\mathrm{T}}=69$ MeV.

The wave function of the three-body system may be written in the resonating group form

$$\left.\begin{aligned} \Psi(1,2,3) &= (1-P_{12})\psi(1,2,3), \\ \psi(1,2,3) &= \sum_{i=1}^{6}\chi_0(\mathbf{u})F_0^{\,i}(\mathbf{r})\sigma^i(1,2,3), \end{aligned}\right\} \qquad \ldots\ldots(2)$$

where P_{12} is the operator exchanging all coordinates of 1 and 2 and where if $\mathbf{r}_1$, $\mathbf{r}_2$, $\mathbf{r}_3$ are the position vectors of protons 1 and 2 and neutron 3 respectively,

$\mathbf{u}=\mathbf{r}_2-\mathbf{r}_3$, $\mathbf{r}=-\mathbf{r}_1+\frac{1}{2}(\mathbf{r}_2+\mathbf{r}_3)$. $\chi_0(u)$ the ground state wave function of the deuteron was approximated by the gaussian function

$$\chi_0(u)=(4\pi)^{-1/2}N_s\exp(-\lambda u^2),$$

with $N=\{0{\cdot}295/\Gamma(3/2)\}^{1/2}$, $\lambda=0{\cdot}0726\times10^{26}\,\mathrm{cm}^{-2}$ and $\sigma(1,2,3)$ represents the six possible spin wave functions for elastic scattering listed in I. The wave equation is

$$\left(-\frac{\hbar^2}{M}\nabla_u{}^2-\frac{3}{4}\frac{\hbar^2}{M}\nabla_r{}^2+V(\mathbf{t})+V(\mathbf{u})+V(\mathbf{v})-E\right)\Psi(1,2,3)=0, \quad \ldots\ldots(3)$$

where $\mathbf{v}=\mathbf{r}_3-\mathbf{r}_1$, $\mathbf{t}=\mathbf{r}_1-\mathbf{r}_2$ and E the total energy is connected with the wave vectors $\mathbf{k}_0$, $\mathbf{k}_n$ of the incident proton before and after scattering by the relation $E=4Mk_0{}^2/3\hbar^2=4Mk_n{}^2/3\hbar^2$.

The wave function (2) may be substituted into the wave equation (3) and expressions for the scattering amplitude $f^{tl}(\theta,\phi)$ obtained as described in I.

It is found that the expression for the amplitudes corresponding to quartet and to tensor scattering, does not contain the two-body singlet potentials V^{S}. Born's approximation may be used in calculating these amplitudes, and the calculation follows the course described in I, with the exception that as the radial variation of the potential well is exponential rather than gaussian, a number of the integrations cannot be performed analytically, but have to be computed numerically.

The expression for the doublet amplitude, on the other hand, depends in the main on the singlet two-body potential V^{S}. It may be written

$$f^{\mathrm{D}}(\theta,\phi)=-(M/3\pi\hbar^2)[\tfrac{9}{8}(L_1{}^{\mathrm{S}}+L_2{}^{\mathrm{S}})+\tfrac{1}{8}(L_1{}^{\mathrm{T}}+L_2{}^{\mathrm{T}})-\tfrac{1}{2}L_3{}^{\mathrm{T}}], \quad \ldots\ldots(4)$$

where the integrals L are defined by

$$\left.\begin{aligned}
L_1{}^{\mathrm{S}}&=\int\chi_0{}^2(u)V^{\mathrm{S}}(t)\exp(-i\mathbf{k}_n\cdot\mathbf{r})F_0(\mathbf{r})\,d\mathbf{r}\,d\mathbf{u},\\
L_2{}^{\mathrm{S}}&=\int\chi_0(u)\chi_0(v)V^{\mathrm{S}}(t)\exp(-i\mathbf{k}_n\cdot\mathbf{r})F_0(\mathbf{r}')\,d\mathbf{r}\,d\mathbf{u},\\
L_1{}^{\mathrm{T}}&=\int\chi_0{}^2(u)V^{\mathrm{T}}(t)\exp(-i\mathbf{k}_n\cdot\mathbf{r})F_0(\mathbf{r})\,d\mathbf{r}\,d\mathbf{u},\\
L_2{}^{\mathrm{T}}&=\int\chi_0(u)\chi_0(v)V^{\mathrm{T}}(t)\exp(-i\mathbf{k}_n\cdot\mathbf{r})F_0(\mathbf{r}')\,d\mathbf{r}\,d\mathbf{u},\\
L_3{}^{\mathrm{T}}&=\int\chi_0(u)\chi_0(v)V^{\mathrm{T}}(u)\exp(-i\mathbf{k}_n\cdot\mathbf{r})F_0(\mathbf{r}')\,d\mathbf{r}\,d\mathbf{u},
\end{aligned}\right\} \quad \ldots\ldots(5)$$

where $\mathbf{r}'=-\mathbf{r}_2+\frac{1}{2}(\mathbf{r}_1+\mathbf{r}_3)$.

In Born's approximation, $F_0(\mathbf{r})$ is replaced by a plane wave $\exp(i\mathbf{k}_0\cdot\mathbf{r})$. This replacement cannot now be made in view of the singularity in V^{S} at small values of t. Even if a non-infinite potential were considered, it would be impossible to reduce the product r_0V^{S} to such a value that Born's approximation might be applied, without destroying the agreement with the p–p scattering data.

It may be observed that (*a*) as the deuteron has an 'extended structure' and as the radius of the repulsive core ($0{\cdot}6\times10^{-13}$ cm) is small, each scattering centre in the deuteron may be considered to act independently at high enough energies, at least as far as the core is concerned; (*b*) the main contribution to the integrals L_1 and L_2 derives from regions where $|\mathbf{t}|$ is small. With these considerations

in mind, it was decided to approximate the functions $F_0(\mathbf{r})$ by $F_0'(\mathbf{t})$, and $F_0(\mathbf{r}')$ by $F_0'(-\mathbf{t})$, in the doublet integrals L, where

$$F_0'(\mathbf{t}) = \sum_{n=0}^{\infty} (2n+1) i^n \exp(i\eta_n) L_n(t) P_n(\cos\theta); \qquad \ldots\ldots(6)$$

$\cos\theta = (\mathbf{k}_0 \cdot \mathbf{t}, k_0 t)$, and $L_n(t)$ and η_n are the radial wave functions and phase shifts for scattering by an impenetrable sphere of radius r_0 (Mott and Massey 1949)

$$\left.\begin{aligned} L_n(t) &= (\tfrac{1}{2}\pi k_0 t)^{1/2}[\cos\eta_n J_{n+1/2}(k_0 t) + (-1)^n \sin\eta_n J_{-n-1/2}(k_0 t)], \quad t > r_0 \\ &= 0, \qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\quad t < r_0 \end{aligned}\right\} \quad \ldots\ldots(7)$$

and $$\eta_n = \tan^{-1}[(-1)^{n-1} J_{n+1/2}(k_0 r_0)/J_{-n-1/2}(k_0 r_0)].$$

For large $\mathbf{r}$ and $\mathbf{t}$ $F_0'(t)$ has the correct asymptotic form

$$F_0'(t) \propto \exp(ik_0 z) + r^{-1}\exp(ik_0 r) f(\theta, \phi).$$

Substituting the expressions (6) and (7) into the integrals (5) and changing the variables of integration from $\mathbf{r}, \mathbf{u}$ to $\mathbf{t}, \mathbf{u}$, the integrations over the angular variables and over u may be performed analytically, and the remaining integration over t can be carried out numerically. In practice it proved sufficient to take only the first four or five terms in the series (6).

To obtain some estimate of the size of error arising from the above approximation, the singlet p–p scattering cross section has been computed at 340 Mev, by replacing $F_0(\mathbf{r})$ in the matrix element $\int \exp(-i\mathbf{k}_n \cdot \mathbf{r}) V(r) F_0(\mathbf{r})\, d\mathbf{r}$ by $F_0'(\mathbf{r})$ and fair agreement has been found with the results of Jastrow, as is seen from the table.

Angle in c.m. system (°)	0	10	20	40	60	80	90
$A \times 10^{-27}$ cm²	9·2	7·8	5·9	2·0	1·0	3·1	3·6
$B \times 10^{-27}$ cm²	8·5	7·4	4·3	0·7	1·9	3·0	3·3

A: calculated by replacing $F_0(\mathbf{r})$ by $F_0'(\mathbf{r})$; B: Jastrow's results at 340 Mev.

§2. NUMERICAL RESULTS

The differential and total elastic cross sections for proton energies of 150 and 240 Mev were computed in the manner outlined above. The differential cross sections are compared in figs. 1 and 2 with the experimental data of Pickavance *et al.* (1951) and Schamberger (1951). The calculated total cross sections are: fig. 1, at 150 Mev, $\sigma_T = 3{\cdot}4 \times 10^{-26}$ cm², fig. 2, at 240 Mev, $\sigma_T = 0{\cdot}97 \times 10^{-26}$ cm².

The largest contribution to the cross section comes from the integrals L_1^S and L_2^S which contain the singlet two-body potentials. The quartet scattering amplitude contributes significantly in the forward direction only, accounting for a third of the cross section at 0°.

Comparison of the experimental and calculated cross sections shows that in (*a*) the calculated scattering near 90° is too large, but shows a flattening of the cross section similar to that indicated by the experimental data; (*b*) experimentally there is no evidence for the maximum at 180° in the centre-of-mass system shown by the calculated results and the calculated forward scattering is seen to be too small; (*c*) if Schamberger's differential cross section is integrated over the scattering angles, the experimental total cross section at 240 Mev is, at the least, of the order of $1{\cdot}2 \times 10^{-26}$ cm². This is of the same order of magnitude as the calculated cross section of $0{\cdot}97 \times 10^{-26}$ cm², but the total cross section at 150 Mev

appears to be too large to be consistent with the experimental data, the total cross section at 95 MeV being about $2{\cdot}4\times10^{-26}\,\mathrm{cm}^2$ (estimated by Pickavance *et al.* 1951, from the data of Stern and Bloom 1951).

It is of interest to note that the cross sections calculated using potentials without a repulsive core (I, Wu and Ashkin 1948, Chew 1948, Auerbach (reported by Schamberger 1951)), are in error in the opposite sense to (*a*) and (*b*), the scattering in the forward direction being too large and the cross section at 90° being far too small. The approximation used in finding the doublet amplitude may considerably overestimate the effect of the repulsive core for the following reason. Consider the integral $L_1{}^S$ which is the most important factor at all angles. If the core is absent and Born's approximation is made, then $L_1{}^S$ contains as a factor $\int d\mathbf{u}\chi_0{}^2(u)\exp\{i(\mathbf{k}_0-\mathbf{k}_n)\cdot\mathbf{u}/2\}$. This depends only on the

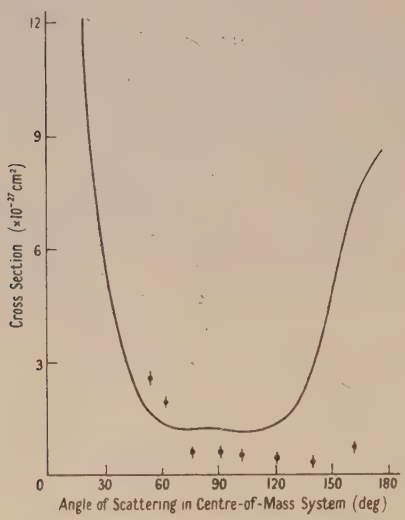

Fig. 1. Comparison of calculated and experimental angular distributions for n–d scattering at 150 MeV. The experimental points are taken from Pickavance *et al.* (1951).

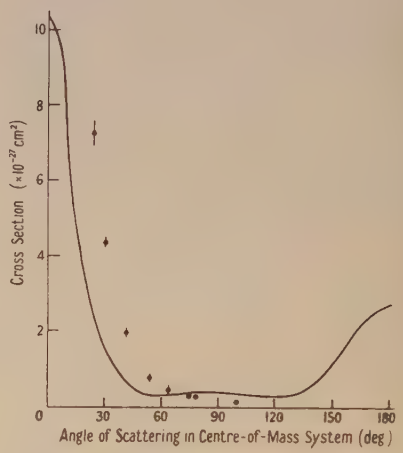

Fig. 2. Comparison of calculated and experimental angular distributions for n–d scattering at 240 MeV. The experimental points are taken from Schamberger (1951).

ground state wave function of the deuteron and on the momentum transfer $|\mathbf{k}_0-\mathbf{k}_n|$. With $\chi_0(u)=(4\pi)^{-1/2}N_S\exp(-\lambda u^2)$ it is proportional to

$$\exp(\alpha|\mathbf{k}_0-\mathbf{k}_n|^2).$$

Even if Born's approximation is not made, Chew (1948, 1950, 1951) has shown that the scattering amplitude contains a 'sticking factor' of this type at small angles; at large angles this may not be true as the scattering is then of the three-body 'pick-up' type that has been discussed by Chew and Goldberger (1950). Making the approximation of replacing $F_0(\mathbf{r})$ by $F_0'(\mathbf{t})$, the corresponding factor for L^S is $\int d\mathbf{u}\chi_0{}^2(u)\exp(-i\mathbf{k}_n\cdot\mathbf{u}/2)$ which is proportional to $\exp(-\alpha k^2)$ and is independent of angle. This approximation would be expected therefore to overestimate the scattering at large angles and underestimate that in the forward direction.

It cannot definitely be stated whether or not Jastrow's interaction is capable of explaining the three-body collision problem, but it is encouraging that the

calculated scattering around 90° is of the correct order of magnitude and shows a flattening similar to that indicated by the experimental results and therefore represents a definite improvement over the previous calculations using non-singular potentials.

To conclude, it appears that more refined, and of necessity lengthy, calculations are needed in applying Jastrow's or similar interactions to three-body problems and until the two-body potentials are known with more certainty it would not seem profitable to attempt this work.

ACKNOWLEDGMENTS

I should like to thank Professor H. S. W. Massey for suggesting this problem and Dr. E. H. S. Burhop for his interest and for some extremely helpful correspondence.

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Coherent Scattering of Gamma-Rays by Bound Electrons

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Communicated by R. E. Peierls; MS. received 16th June 1952

ABSTRACT. The coherent scattering of high energy gamma-rays by bound electrons in light atoms is investigated. The bound intermediate states of the electron are expanded in terms of free states and potential scatterings. It is found that not only the zero-order term of this expansion, but also the first-order term, must be kept, and that the latter is actually the larger of the two for large momentum transfer.

§ 1. INTRODUCTION

We have made an estimate of the amplitude for coherent scattering of high-energy gamma-rays ($\hbar\omega > 1$ Mev) by light atoms. Contrary to what might be expected, we conclude that it is essential to include both the zero-order and first-order terms in the Born expansion of intermediate states. In other words, one must take into account the binding of the electron in the intermediate state to first order. Thereafter the subsequent terms seem to decrease as successive powers of $Z\alpha$.

§ 2. DERIVATION

We follow the notation and methods of Feynman (1949) and deal only with scattering by one K-shell electron in an atom. We consider the scattering of photons with momentum $\mathbf{q}$, energy q, polarization vector $\mathbf{e}$, the emitted photons having momentum $\mathbf{q}'$, energy q and polarization vector $\mathbf{e}'$, such that both q and $\Delta q\,(=|\mathbf{q}-\mathbf{q}'|)$ are large compared with m, the rest mass of the electron. The zero-order term in this Born expansion of intermediate states, after the neglect of various relativistic effects, gives the usual form-factor (Franz 1935), which is (using the Dirac wave function for one electron)

$$F_0=\frac{Z\alpha m\sin\{2\gamma\tan^{-1}(\Delta q/2Z\alpha m)\}}{\gamma\Delta q\{1+(\Delta q/2Z\alpha m)^2\}^{\gamma}}, \qquad \ldots\ldots(1)$$

where $\gamma=(1-Z^2\alpha^2)^{1/2}$. This reduces, for $\Delta q\gg Z\alpha m$ and $Z\alpha\ll 1$, to

$$F_0\simeq 2\pi\left(\frac{Z\alpha m}{\Delta q}\right)^3 Z^2\alpha^2\left(1+\frac{8m}{\pi Z\alpha\Delta q}\right). \qquad \ldots\ldots(2)$$

The quantity occurring naturally in our perturbation calculation is the transition matrix element, of which the zero-order term A_0 is related to the form-factor by

$$A_0=(2\pi e^2/mq)F_0(\mathbf{e}\,.\,\mathbf{e}'). \qquad \ldots\ldots(2\,a)$$

The relativistic terms neglected in the derivation of (1) have been discussed by Levinger (1952 and private communication), and his results show that their effect is to increase F_0 by a quantity of order $Z\alpha(\Delta q/m)F_0$ when $\Delta q\leqslant m$. The corresponding corrections when $\Delta q\gg m$ are not yet known.

We perform the calculation for a K electron only, because the K shell contributes the largest part of the scattering. One of us (J. B. W.), as well as Rohrlich and Rosenzweig (Levinger, private communication), has calculated the form-factor for the whole of the L shell and estimated that the zero-order contribution would be about one-eighth that of the K shell for large values of Δq.

The first-order term is obtained by using $K_+^{(1)}(2,1)$ (Feynman 1949, eqn. (13)) in place of $K_+(2,1)$ which was used in obtaining A_0. [In this notation $K_+(2,1)$ may be defined by $K_+(2,1)=(i/16\pi^4)\int(\boldsymbol{p}-m)^{-1}\exp[-i(p_\mu x_{1\mu}-p_\mu x_{2\mu})]\,d^4p$, where $\boldsymbol{p}$ stands for $p_\mu\gamma_\mu$, with the summation convention $a_\mu b_\mu=a_4b_4-a_1b_1-a_2b_2-a_3b_3$. The other symbols in bold italics throughout the paper are similarly defined. Our γ_μ differ from Pauli's by a factor i for $\mu=1,2,3$. In eqn. (4) we shall make use of the three-dimensional Fourier transform $Ze^2\gamma_4/(2\pi^2\mathbf{k}^2)$ of the electrostatic potential $\boldsymbol{V}=Ze^2\gamma_4/r$. Hence $\boldsymbol{k}$ will only have components with suffices 1, 2 and 3. $\boldsymbol{p}$, $\boldsymbol{q}$ and $\boldsymbol{q}'$ involve the momentum components and energies of the bound electron in its initial state, the incident photon and the emitted photon respectively.] As a result

$$A_1=\int d^3x''\,d^4x'\,d^4x\bar{\psi}(x'')\boldsymbol{A}'(x'')K_+(x''-x')\boldsymbol{V}(x')K_+(x'-x)\boldsymbol{A}(x)\psi(x)$$
$$+\text{term with interchange of photon operators.} \qquad \ldots\ldots(3)$$

Here $\boldsymbol{A}(x)$ is the vector potential of the incoming photon and $\boldsymbol{A}'(x'')$ that of the outgoing. In terms of momentum variables

$$A_1=-(8\pi^2Ze^4/q)\int d^3p\,d^3k\bar{\psi}(\mathbf{p}+\Delta\mathbf{q}+\mathbf{k})[\mathbf{e}'(\boldsymbol{p}+\boldsymbol{q}+\boldsymbol{k}-m)^{-1}\gamma_4\mathbf{k}^{-2}(\boldsymbol{p}+\boldsymbol{q}-m)^{-1}\mathbf{e}$$
$$+\mathbf{e}(\boldsymbol{p}-\boldsymbol{q}'+\boldsymbol{k}-m)^{-1}\gamma_4\mathbf{k}^{-2}(\boldsymbol{p}-\boldsymbol{q}'-m)^{-1}\mathbf{e}']\psi(\mathbf{p}). \qquad \ldots\ldots(4)$$

It is important to note that p_4, which appears in the denominators of (4), is just E_0, the total energy of the bound electron, and this gives rise to the possibility of these denominators vanishing.

On account of the singularities the integral is defined by replacing m in (4) by $m-i\epsilon$, where ϵ is small. Then, in addition to the principal value of the integral, there is an imaginary part which is obtained by taking π times one-half of the residues. This represents coherent scattering which is 90° out of phase with the other coherently scattered radiation, and is due to absorption (cf. Rohrlich and Gluckstern 1952). The amplitude for this absorption scattering in the case of the zero-order term has been estimated by Levinger (private communication) for $\Delta q \leqslant m$, and he finds it to be negligible. One can see that for high values of Δq the residues will be small in the zero- and first-order terms, as they involve initial and final wave functions of high momenta. We neglect these contributions in what follows and take only principal values of the integrals.

The expression (4) represents the single scattering of the electron by the potential and therefore includes the possibility of the electron picking up from the potential enough momentum $\mathbf{k}(\simeq -\Delta\mathbf{q})$ to compensate approximately for the momentum transfer to the electromagnetic field. Indeed, the regions in which the wave functions, rather than V, furnish the momentum difference $\Delta\mathbf{q}$ give a negligible contribution, as can be seen immediately by inspection of the relativistic momentum transforms of the wave functions. In the important region of the integration in (4) the magnitudes of $\mathbf{p}$ and $\Delta\mathbf{q}+\mathbf{k}$ are of order $Z\alpha m$, and these quantities can be neglected compared with $\mathbf{q}$ or $\mathbf{q}'$. For instance, $(\boldsymbol{p}+\boldsymbol{q}-m)^{-1}=(\boldsymbol{p}+\boldsymbol{q}+m)/[(\boldsymbol{p}+\boldsymbol{q})^2-m^2]\simeq \boldsymbol{q}/2mq$, $\boldsymbol{p}+m$ going out since $\boldsymbol{p}$ anti-commutes with $\boldsymbol{e}$ except for velocity dependent effects, and $\boldsymbol{p}-m$ gives only negligible v^2/c^2 effects when applied to the wave function. The term in square brackets in (4) then becomes

$$(\boldsymbol{e}'\boldsymbol{q}'\gamma_4\boldsymbol{q}\boldsymbol{e}+\boldsymbol{e}\boldsymbol{q}\gamma_4\boldsymbol{q}'\boldsymbol{e}')/4m^2q^2k^2, \qquad \ldots\ldots(5)$$

which reduces immediately to

$$\gamma_4(2m^2k^2)^{-1}\Theta, \qquad \ldots\ldots(6)$$

where the angle-dependent factor is

$$\Theta=q^{-2}[(\mathbf{e}\,.\,\mathbf{e}')(q^2+\mathbf{q}\,.\,\mathbf{q}')-(\mathbf{e}'\,.\,\mathbf{q})(\mathbf{e}\,.\,\mathbf{q}')]. \qquad \ldots\ldots(6\,a)$$

Thus

$$A_1\simeq -4\pi^2Ze^4m^{-2}q^{-1}\Theta\int d^3p\,d^3k\bar{\psi}(\mathbf{p}+\Delta\mathbf{q}+\mathbf{k})\gamma_4\mathbf{k}^{-2}\psi(\mathbf{p}). \qquad \ldots\ldots(7)$$

As pointed out above, the only significant contributions to (7) come from regions where $|\mathbf{p}+\Delta\mathbf{q}+\mathbf{k}|$ and $|\mathbf{p}|$ are of order $Z\alpha m$. Consequently, non-relativistic wave functions can be used with negligible error, but the integration should, in principle, extend only up to values of $|\mathbf{p}+\Delta\mathbf{q}+\mathbf{k}|$ and $|\mathbf{p}|$ of the order of 5 or 10 times $Z\alpha m$. Beyond this region, however, the non-relativistic functions cut off the contributions strongly, contributing by a factor $(Z\alpha m/\Delta q)^2$ less than in the important region. Hence the integration can just as well be extended to infinity. With these approximations one finds

$$A_1\simeq -4\pi e^2m^{-1}q^{-1}Z^2\alpha^2(Z\alpha m/\Delta q)^2\Theta. \qquad \ldots\ldots(8)$$

We see that the ratio of (8) to (2 *a*), apart from angle dependence and sign, is

$$\frac{1}{\pi}\left(\frac{\Delta q}{Z\alpha m}\right)\left(1+\frac{8m}{\pi Z\Delta q}\right)^{-1}. \qquad \ldots\ldots(9)$$

In fact this ratio is unity when $\Delta q\simeq 2\sqrt{2}m$ and increases with Δq. Consequently the first-order term (8) is the main term in most of the photon energy range under consideration, since subsequent terms will involve higher powers of $Z\alpha$.

The momentum transferred to the photon must in any event be supplied by the nucleus through its coulomb field. In the zero-order term this proceeds through the coulomb forces which act while the electron is in the ground state. We see, from the first-order term, that the dominant process is one in which most of the momentum balance is transferred to the nucleus between absorption and re-emission (or vice versa) of the photon. Our result about the relative magnitudes of the two terms means then that the transfer of a large amount of momentum is easier when the electron already has a large momentum than when it is slow.

ACKNOWLEDGMENTS

We wish to thank Professor R. E. Peierls for many helpful discussions. One of us (J. B. W.) acknowledges with thanks a grant from the Department of Scientific and Industrial Research.

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Angular Distribution of γ-Radiation following a Deuteron Stripping Reaction

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MS. received 20th June 1952

ABSTRACT. Expressions are derived theoretically for the angular distribution of γ-radiation from a deuteron stripping reaction, measured in coincidence with a fixed direction of the emitted particle.

Theoretical treatment of (d, p) and (d, n) stripping reactions by Butler (1951) and Bhatia *et al.* (1952) have shown that the angular distribution of the emitted particle enables the orbital angular momentum of the captured particle to be determined, and hence gives information concerning the spin of the nuclear state after capture, if that of the initial nucleus be known.

If the state after capture is an excited state, further information may be derived from the angular distribution of the emitted γ-radiation measured in coincidence with a fixed direction of the emitted particle, this direction being chosen to coincide with a peak of the particle angular distribution.

Expressions for the γ-ray distribution are derived below from a simple formulation of stripping theory.

Consider a (d, p) reaction for definiteness. The wave function of the incident deuterons in the centre-of-mass system*

$$\psi_{\mathbf{k}_d\mu_d}(\mathbf{r}_n, \sigma_n, \mathbf{r}_p, \sigma_p) = \exp[i\mathbf{k}_d \cdot (\mathbf{r}_n + \mathbf{r}_p)/2]\psi(|\mathbf{r}_n - \mathbf{r}_p|)C^1_{\mu_d\,\sigma_n^{\frac{1}{2}}\sigma_p^{\frac{1}{2}}} \quad \ldots\ldots(1)$$

* The notation of Bhatia *et al.* (1952) is used throughout except where specified. The C's are Wigner coefficients used in the theory of addition of quantized angular momenta (Racah 1942). The suffixes are omitted for typographical convenience in eqns. (2) to (4).

can be expressed as a superposition

$$\Sigma_{\mathbf{k}_p} \exp(i\mathbf{k}_p \cdot \mathbf{r}'_p)\Phi_{\mathbf{k}_p}(\mathbf{r}_n)C \quad \ldots\ldots(2)$$

in each term of which the proton has definite linear momentum $\mathbf{k}_p$. Multiplying both sides by $\exp(-i\mathbf{k}_p \cdot \mathbf{r}'_p)$ and integrating over $\mathbf{r}_p$, we obtain $\Phi_{\mathbf{k}_p}(\mathbf{r}_n) = \exp(i\mathbf{k} \cdot \mathbf{r}_n)G(K)$ where $\mathbf{k} = \mathbf{k}_d - (M_i/M_f)\mathbf{k}_p$, $K = |\mathbf{k}_p - \mathbf{k}_d/2|$, and G is the function given by Bhatia *et al.* (1952), eqn. (17). Thus

$$\psi_{\mathbf{k}_d\mu_d} = \Sigma_{\mathbf{k}_p} G(K) \exp(i\mathbf{k}_p \cdot \mathbf{r}'_p) \exp(i\mathbf{k} \cdot \mathbf{r}_n)C. \quad \ldots\ldots(3)$$

The stripping assumption is that in the state after capture resulting from each term of the superposition (3), $\mathbf{k}_p$ and σ_p are unchanged. Since the proton momentum $\mathbf{k}_p$ is observed, only one state of the superposition (3) can be effective in the transition, and in that state the incident neutron behaves simply as a plane wave travelling in the direction $\mathbf{k}$.

Expansion of the neutron plane wave into spherical waves about a polar axis in the direction $\mathbf{k}$ leads to

$$\psi_{\mathbf{k}_d\mu_d} = \Sigma_{\mathbf{k}_p} \exp(i\mathbf{k}_p \cdot \mathbf{r}'_p) C \Sigma_{l_n} g^0_{l_n}(r_n) Y^0_{l_n}(\theta_n \phi_n) \quad \ldots\ldots(4)$$

where $g^0_{l_n}(r_n)$ is the function defined by Bhatia *et al.* (1952), eqn. (16), with r_n written for R.

The further assumption that the probability of capture from each l_nth partial wave is proportional to the 'penetrability' of that wave at $r_n = R$ (i.e. to $|g^0_{l_n}(R)|^2$) multiplied by a constant of proportionality, leads at once to the result of Bhatia *et al.* (1952). The proportionality constant ("intrinsic probability of capture of neutrons reaching the nuclear surface with orbital angular momentum l_n") corresponds except for numerical factors to the quantity Λ_{l_n} of Bhatia *et al.* (1952). Matching of the incident neutron wave at the surface of the nucleus would lead to the more complicated result of Butler (1951).

However, the γ-ray angular distribution does not depend on the capture probability, but only on the relevant angular momenta, and is the same as for capture from a beam of incident neutrons in the direction $\mathbf{k}$.

The wave function of the system deuteron plus initial nucleus $\psi_{\mathbf{k}_d\mu_d}\chi_{J_iM_i}$ may be written, using (4), as

$$\Sigma_{\mathbf{k}_p} \exp(i\mathbf{k}_p \cdot \quad) \Sigma_{J_eM_e} \Sigma_{l_n j_n \mu_n} \{C^{1\ \frac{1}{2}\ \frac{1}{2}}_{\mu_d\ \mu_n\ \sigma_p} C^{j_n\ l_n\ \frac{1}{2}}_{\mu_n\ 0\ \mu_n} C^{J_e\ \ J_i\ j_n}_{M_e\ M_i\ \mu_n}\} \psi_{J_eM_e j_n l_n} \quad \ldots\ldots(5)$$

where $j_n = l_n \pm \frac{1}{2}$ is the total angular momentum (spin plus orbital) of the neutron, and $\psi_{J_eM_e j_n l_n}$ is a state of the system neutron plus initial nucleus with total angular momentum J_eM_e.

Assuming that capture leads to an excited state with only one value of J_e and that only one value of l_n contributes (γ's in coincidence with protons in direction $\mathbf{k}_p$ giving a distinct peak), also one value of j_n,* the weighing factor for the orientations M_e of the excited state is given by the square modulus of the expression in curly brackets in eqn. (5). Assuming pure 2^L multipole emission to a state of spin J_f, the γ angular distribution $I(\theta)$ about the axis $\mathbf{k} = \mathbf{k}_d - (M_i/M_f)\mathbf{k}_p$ is readily obtained by multiplying the angular distribution of γ-radiation from each orientation M_e by this weighting factor and summing the resulting expression

* This seems more in keeping with the shell model than the usual procedure of combining first the spins of the initial particles then forming the resultant of total spin plus orbital angular momentum.

over μ_d, M_i, σ_p. Using the orthogonality properties of Wigner coefficients, the expression simplifies to

$$I(\theta) = \Sigma_{M_i \mu_n} |C^{J_e\ J_i\ j_n}_{M_e\ M_i\ \mu_n}|^2 \Sigma_{M_f} |C^{J_e\ J_f\ L}_{M_e\ M_f\ M}|^2 F_{LM}(\theta) \qquad \ldots\ldots(6)$$

where $M = M_e - M_f$, $M_e = M_i + \mu_n$, $\mu_n = \pm\frac{1}{2}$, and the 2^L pole angular distributions $F_{LM}(\theta)$ are tabulated, e.g. by Arnold (1950).

The highest power of $\cos^2\theta$ in $I(\theta)$ is $\cos^{2p}\theta$, where p is the greatest integer less than or equal to j_n, J_e, L and hence $I(\theta)$ is isotropic if $j_n = \frac{1}{2}$ or $J_e = 0$ or $\frac{1}{2}$. Note that with the assumptions made the result (6) is free of adjustable parameters, and is independent of l_n except in so far as j_n can only have one of the two values $l_n \pm \frac{1}{2}$.

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Some Absolute Determinations of Proton–Gamma Resonances in the Energy Range below 500 kev

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Communicated by D. R. Chick; *MS. received* 1*st November* 1951, *and in amended form* 26*th March* 1952

ABSTRACT. An electrostatic analyser of high accuracy has been used to measure and control the energy of a proton beam accelerated by an electrostatic generator. By using a deflecting angle of $\pi/2\sqrt{2}$ radians, a wide incident beam was focused on to a narrow exit slit.

Target currents of 10 μA were obtained homogeneous in energy to one part in a thousand. The beam energies were also measured absolutely to this accuracy.

The instrument was used to determine the energy of the ^{7}Li(p, γ) resonance, which was found to be $441{\cdot}5 \pm 0{\cdot}5$ kev with a half width of $12{\cdot}2 \pm 0{\cdot}5$ kev. The energies of two sharp resonances in fluorine were found to be $340{\cdot}4 \pm 0{\cdot}4$ and $483{\cdot}1 \pm 0{\cdot}5$ kev, with half widths $2{\cdot}9 \pm 0{\cdot}2$ kev and $2{\cdot}2 \pm 0{\cdot}2$ kev respectively.

§1. INTRODUCTION

Absolute determinations of (p, γ) resonances below 500 kev have been made by several investigators using resistance voltmeters. In general, accuracies within 1% are claimed for this method, although Heydenburg is reported to have obtained much higher accuracies than this (Morrish 1949). Other workers also claimed higher accuracies for measurements relative to an absolute determination of the fluorine resonance at 873·5 kev which was made by Herb, Snowden and Sala (1949) using an electrostatic analyser, whilst Morrish has used an electrostatic analyser calibrated by a low-energy electron beam.

The instrument described in the present work gave an absolute measurement of beam energy in terms of its geometric dimensions and the potential difference across the electrodes. Accurate measurement of these reduced the error to

less than one part in a thousand. The source of high energy particles was an air insulated electrostatic generator. The instrument was also used to control the energy of the particles by intercepting a fraction of the beam on each jaw of an exit slit. Any inequality in the currents on the slit jaws constituted an 'error signal' which was fed back to correct the voltage of the generator so as to return the beam to the median plane of the analyser. The control mechanism has been described by Chick and Inall (1952).

A deflecting angle of $\pi/2\sqrt{2}$ radians focused a wide parallel incident beam on to a narrow exit slit, obtaining a resolution of one part in a thousand with target currents of up to 10 μA. This compares favourably with currents obtained using 90° electrostatic analysers of comparable resolution (Warren, Powell and Herb 1947).

§ 2. SUMMARY OF THEORY

The relation between the potential difference V through which a charged particle has fallen and the potential difference X between the electrodes is approximately

$$2V = X(R_1 R_2)^{1/2}/(R_2 - R_1) \qquad \ldots\ldots(1)$$

where R_2 and R_1 are the radii of the electrodes ($R_2 > R_1$). The error involved is small for small $R_2 - R_1$, and in the present case where $(R_2 - R_1)/(R_1 R_2)^{1/2} \simeq 0{\cdot}02$ it was less than one part in 10^4. The potential through which the particle had fallen was, therefore, calculated to the required accuracy by careful measurement of X, $R_2 - R_1$ and $(R_1 R_2)^{1/2}$.

Hertzog (1934) has determined the following focusing condition for a beam of particles describing a circle of radius a in a radial electrostatic field: $(l' - g)(l'' - g) = f^2$ where l' = object distance measured from the entrance plane of the analyser, l'' = image distance measured from the exit plane of the analyser, $g = a/\sqrt{2} \cot \theta\sqrt{2}$ = distance of focal point from the exit plane of the analyser, $f = a/\sqrt{2} \operatorname{cosec} \theta\sqrt{2}$ = focal length, θ = angle of deflection. By choosing the deflecting angle to make $g = 0$ ($\theta = \pi/2\sqrt{2}$), and placing the exit slit at the point $l'' = 0$, it can be shown that for a parallel incident beam the energy resolution is independent of the width of the entrance slit to a first order of approximation. This ideal condition was approached sufficiently closely to allow an energy resolution of 1000 to be obtained without restricting the width of the entrance slit to less than the electrode separation.

§ 3. EXPERIMENTAL METHOD

(i) *Description of Apparatus*

A photograph of the analyser is shown in fig. 1. The mean radius of the deflector electrodes was 12 in. and the nominal width of the gap was 0·25 in. The electrodes were of mild steel and were thoroughly stress-relieved before final machining. After the final machining, the separation was constant to somewhat better than 0·0001 in. (fig. 2).

The entrance slit was fixed with a separation of $\frac{3}{16}$ in. in order to prevent bombardment of the analyser electrodes by the beam. The exit slit assembly consisted of two pairs of slit jaws, one fixed with a separation of 0·006 in. corresponding to an energy resolution of 1000, the error signal being taken from these slit jaws, the other mounted immediately behind these 'signal slits' and adjustable to enable resolution to be increased up to one part in ten thousand.

(ii) *Deflecting Voltage Supplies*

A block diagram of the voltage stabilizing and measuring circuit is shown in fig. 3. The output voltage was controlled by means of a highly stabilized reference voltage source fed from thermostatically controlled dry cells. The reference voltage source supplied up to ± 200 volts and the required fraction of this output was fed into the main potential dividers through R_3. The polarity was opposite to that of the voltage fed into the high potential end, and the ratio

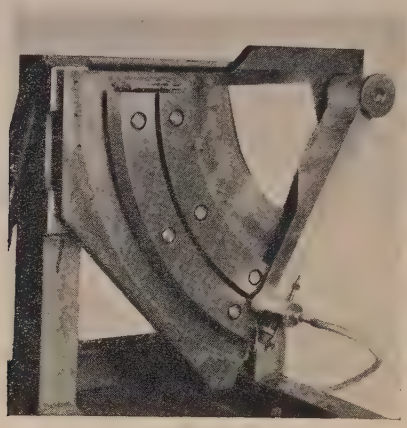

Fig. 1. Photograph of analyser.

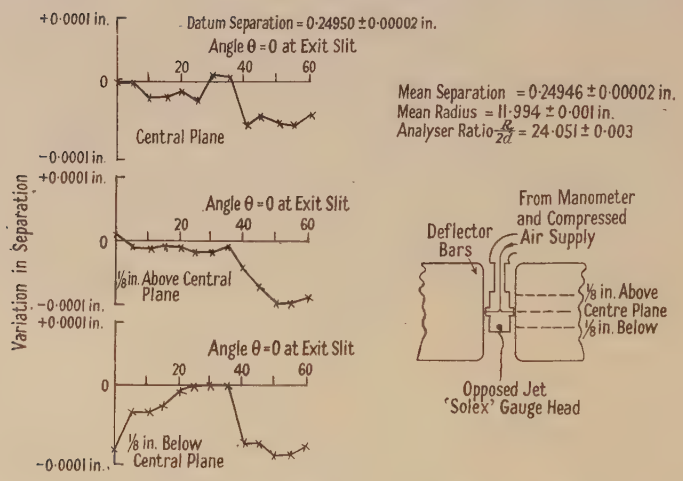

Fig. 2. Variation in deflector electrode separation against angle, over depth occupied by the beam.

R_1/R_3 was such that the points A and A′ were at earth potential. Any drifts in the outputs of the 12 kv supplies resulted in the points A and A′ drifting away from earth potential. The variations in the potentials of A and A′ were fed into d.c. amplifiers and hence to the grids of valves in series with the main h.t. supplies via radio-frequency links. The series valves therefore acted as impedances which varied so as to keep the potentials applied to the dividers and deflector electrodes constant. Rough adjustment on the input to the h.t. power packs

kept their output within the range which could be stabilized by the series valve. The voltage across the low ratio arm of the potential divider was compared with that of standard cells by means of a precision potentiometer. The cells were maintained at a constant temperature of 30° c.

The main potential dividers consisted of sub-units of wire wound resistors enclosed in corona shields and mounted on Perspex insulation. The corona shields were maintained at the mean voltage of the enclosed resistances by an auxiliary chain of carbon resistors. The whole assembly was normally surrounded by aluminium shields to minimize the effects of draughts. Exhaustive tests both on the resistances making up the dividers and on the dividers themselves under actual working conditions enabled the potential

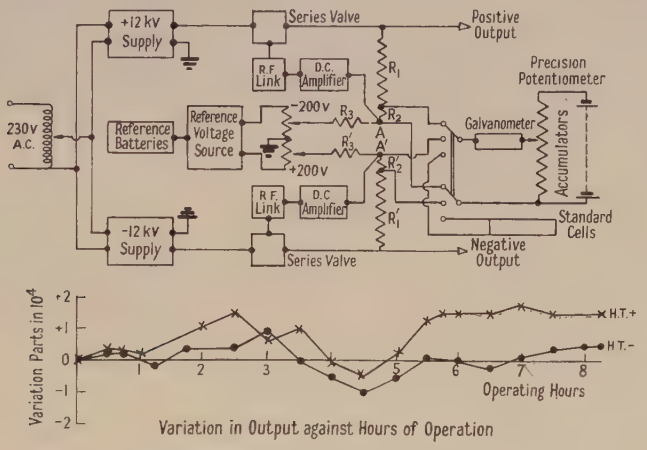

Fig. 3. Block diagram of deflector voltage stabilizing circuit, and plot of measured stability of output.

divider ratio to be estimated with an accuracy to one part in ten thousand after approximately one hour, in which time the dividers reached a thermally stable condition. The ratios were measured under actual working conditions by switching a standard resistance into the high resistance arm of one divider and balancing it out in a Wheatstone bridge circuit by adjusting a variable resistance in series with the low resistance arm. The other potential divider was used as the fixed arms of the bridge. The method is similar to that used by Rymer (1952).

Plots of observed output voltage against hours of operation are shown in fig. 3.

(iii) *Operation of the Analyser*

A very slightly divergent incident beam was used in order to produce a focus at the exit slit, which was 0·5 in. from the effective exit plane of the analyser, and target currents of up to 10 μA were obtained. The current to each of the exit slit jaws was approximately 1 μA, which illustrates the sharpness of the focus.

The exit slit assembly was modified before Run 10 (table 1). The signal slit and beam defining slit were replaced by a single fixed slit of separation 0·006 in. corresponding to a resolution of 1000. This eliminated the uncertainty in the calculation of the effective exit plane (Appendix, error 4). The results obtained were not significantly different from those using the more complicated exit slit assembly.

Table 1

Run	Target thickness†	Resonant energies (kev) (corrected for target thickness) Lithium	Fluorine 340 line	Fluorine 483 line	Half widths (kev) (corrected for target thickness) Lithium	Fluorine 340 line	Fluorine 483 line
1	3 kev LiF	442·0 441·3	340·5	—	—	—	—
2	2 kev LiF	441·3 441·6	340·9	483·9 483·6	11 ± 1·0	—	—
3	2 kev LiF	—	340·8	—	—	—	—
4	500 ev LiF	—	340·2	—	—	3·0 ± 1·0	—
5	500 ev LiF	441·8	340·3 340·6	483·0	12·0 ± 1·0	2·8 ± 0·3 3·0 ± 0·5	2·3 ± 0·3
6	500 ev Li_2O	441·5	—	—	12·0 ± 1·0	—	—
7	500 ev LiF	440·9	340·5 340·0	482·6	12·0 ± 0·5	2·9 ± 0·5	2·1 ± 0·2
8	100 ev MgF_2	—	340·0	—	—	2·8 ± 1·0	—
9	2 kev LiF	441·2	340·0	482·6	13·0 ± 1·0	—	—
10	500 ev LiF	441·5	340·5	483·5	12·5 ± 0·5	3·2 ± 0·5	2·2 ± 0·3
11	100 ev LiF	—	339·8 340·5*	482·4 482·7*	—	2·8 ± 0·5	2·0 ± 0·3
12	100 ev MgF_2	—	340·5 340·0*	483·7 482·4*	—	—	—
13	500 ev LiF	441·5	340·7	483·5	12·5 ± 0·5	3·5 ± 0·5	2·2 ± 0·5
14	2·5 kev MgF_2	—	340·5	483·5	—	—	—
15	2·5 kev MgF_2	—	340·6	483·2	—	—	—
Mean	—	441·5	340·4	483·1	12·2 ± 0·5	2·9 ± 0·2	2·2 ± 0·2

* These determinations were made when the analyser was rotated through 180° about the vertical axis.

† For 400 kev incident beam.

(iv) *Targets*

The targets were prepared by evaporating weighed amounts of lithium, lithium fluoride or magnesium fluoride *in vacuo* on to polished copper backings. The energy ΔE lost by the beam in traversing the targets was calculated from published stopping power curves and from Bethe's formula (Livingstone and Bethe 1937).

The true resonant energy was taken as the observed energy of maximum yield minus $\Delta E/2$ (Morrish 1949), and the true half widths Γ_0 were calculated from the observed half widths Γ_1 by applying the formula $\Gamma_1=(\Gamma_0^2+\Delta E^2)^{1/2}$ (Tangen 1946). This correction was used only when ΔE was small compared with Γ_0.

Carbon contamination of the targets due to condensation of diffusion pump oil was avoided by using a liquid nitrogen trap immediately in front of them; the targets were also heated to 250° c during irradiation. The reproducibility of

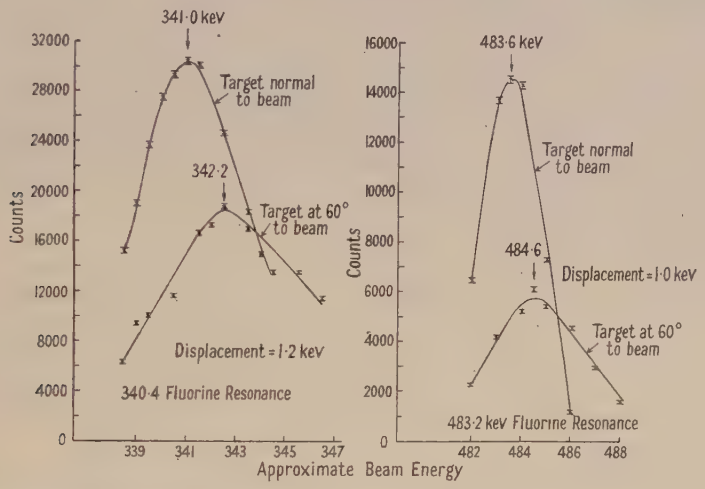

Fig. 4. Displacement of fluorine resonances on rotating a 2 kev lithium fluoride target through 60°.

the results showed that these measures were effective (table 1). The targets were also rotated through 60° thus doubling their effective thickness and the thickness of any surface contamination. The observed displacements confirmed any surface contamination to be negligible, and were used to check the calculated target thickness in energy units ΔE. The agreement was good as is shown in fig. 4. The gamma rays were detected by means of a Geiger–Müller counter with a lead radiator. The background counting rate was reduced by suitable screening to approximately 15 counts/minute and the counter was automatically switched off when the target had received a pre-determined charge.

§ 4. DISCUSSION OF RESULTS

The results obtained in the present work are compared with those of other workers in table 2. Typical curves are shown in fig. 5. The agreement of the present result for the lithium resonance with those obtained by Fowler and Lauritsen and Hudspeth and Swann is satisfactory in that it indicates agreement between the present instrument and the absolute electrostatic analyser of Herb, Snowden and Sala, which has been used over a higher energy range.

The only serious discrepancies occur in the determination of the higher fluorine resonance. The merit can be claimed for the present determination that the resonance was repeatedly cross checked with the lower fluorine resonance and the lithium resonance using the same lithium fluoride targets.

Table 2

Investigator	The lithium resonance: Resonant energy (kev)	Half width (kev)	Method	The fluorine resonances: Resonant energy (kev)	Half width (kev)	Resonant energy	Half width	Method
Hafstad, Heydenburg & Tuve (1936)	440 ±4	—	R. V. (abs.)	324 330	< 4	479	—	R. V. (abs.)
Hanson & Bennedict (1944)	446·5 ±1·5	—	E. D. (abs.)	—	—	—	—	—
Streib, Fowler & Lauritsen (1941)	—	—	—	—	—	479	< 10	G. V.
Tangen (1946)	440 ±2	11 ±2	R. V. (abs.)	339 ±2	1·5	479 ±4	< 2	R. V. (abs.)
Bonner & Evans (1948)	—	14·0	—	—	—	—	—	—
Fowler, Lauritsen & Lauritsen (1948).	—	—	—	338† (339·5)	—	—	—	E. A. relative to Li (440 kev) line.
Heydenburg (Reported by Morrish) (1949)	440·8 ±0·5	—	R. V. (abs.)	339·7 ±0·2	—	—	—	R. V. (abs.)
Fowler & Lauritsen (1949)	441·4 ±0·5	12·0	E. A. rel. to 873·5 kev* F line.	—	—	—	—	—
Morrish (1949)	—	—	—	340·4 ±0·2	3·5	—	—	E. A. (cal. by electron beam).
Hudspeth & Swann (1949)	442·3 ±1·5	—	G. V. rel. to 873·5 kev F line	—	—	—	—	—
Chao, Tollestrupp, Fowler & Lauritsen (1950)	—	—	—	—	—	486	—	E. A. rel. to 873·5 kev line*
Present work	441·5 ±0·5	12·2 ±0·2	—	340·4 ±0·4	2·9 ±0·2	483·1 ±0·5	2·2 ±0·2	E. A. (abs.)

* This result was determined by Herb, Snowden and Sala using an absolute electrostatic analyser.
† Relative value in brackets corrected to refer to their later determination of the lithium resonance.
R. V.=resistance voltmeter ; G. V.=generating voltmeter ; E. D.=electrostatic deflector ; E. A.=electrostatic analyser.

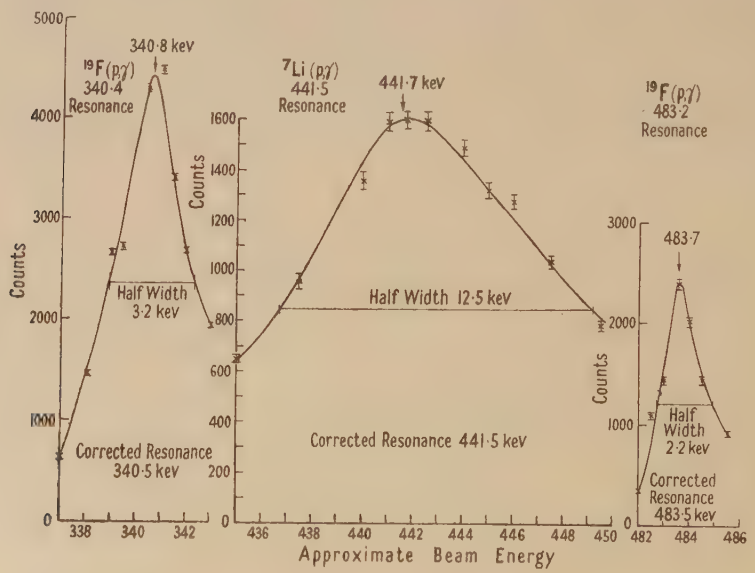

Fig. 5. Resonances observed on a 500 kev thick lithium fluoride target.

ACKNOWLEDGMENTS

The author wishes to thank his colleagues for their helpful suggestions in this work, and particularly D. P. R. Petrie and D. R. Chick for their initial recommendations regarding the design of the analyser, R. Rymer for his assistance in the design of the potential dividers and R. Bailey for his work on the stabilization of the deflector voltages. The late G. O. Jones, in conjunction with Messrs. Wilson & Kyle and our own workshop staff, was responsible for the precision machining of the analyser, and W. M. Jones assisted in the experimental measurement of the nuclear resonances. Thanks are also due to Dr. T. E. Allibone for permission to publish this paper.

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APPENDIX

DISCUSSION OF POSSIBLE ERRORS AND FINAL ACCURACY

(i) *Errors affecting the Determination of the Energy for Maximum Yield*

The nine possible sources of error are discussed below:

1. *Errors in deflector voltage measurement.* The deflector voltage was measured at the peak of each resonance using the circuit shown in fig. 3. Therefore, errors were introduced only by the precision potentiometer and the uncertainty in the ratios of the main potential dividers. The former was certified as being accurate to one part in 10^4, and the main potential divider ratios have been repeatedly checked for a wide range of deflector voltage by the method outlined in § 3 (ii). Their ratios could be estimated to one part in ten thousand.

The total error in the measurement of the deflector voltage was therefore two parts in ten thousand.

2. *Errors in measurement of deflector bar separation.* These measurements were taken by a dynamic air flow method marketed under the trade name Solex. The total separation between the gap faces and those of a slightly undersized slip gauge fitted with radially opposed jets was found by measuring the viscous resistance to air flow of the space between the sides of the gauge and the sides of the gap.

The sensitivity of measurement was 0·000 01 in. and the readings obtained were reproducible to this accuracy. Contours of the gap throughout the depth occupied by the beam are shown in fig. 2.

An absolute measurement of the gap separation was obtained by inserting the gauge head between the jaws of a standard separation gauge, so that the accuracy of the gap measurement became essentially that of the standard gauge, which was $0{\cdot}2500 \pm 0{\cdot}000\,02$ in.

An absolute accuracy to one part in ten thousand can be claimed and measurements made over a period of months were consistent to better than this. All measurements were made at 20° C, but in operation the analyser ratio $(R_2 - R_1)/(R_1R_2)^{1/2}$ is largely independent of temperature.

3. *Radii of deflector bars.* The mean radius was determined at the time of final machining by using clock gauges and the calibrated master rotating plate of a precision Jig Borer. The accuracy of measurement was within 0·0005 in., or four parts in 10^5. The contours of the plates have been checked periodically since, using a single-sided Solex gauge head mounted from a steel arm accurately pivoted at the centre of curvature and playing against each side of the gap in turn. The maximum variation in radius noted was 0·0003 in.

4. *Errors associated with fringing fields at the ends of the analyser.* The fringing fields at the ends of the analyser caused the effective entrance and exit planes to be situated a distance S from the ends of the deflector plates given by

$$S = \frac{2x}{\pi}\tan^{-1}\frac{d}{2x} - \frac{d}{\pi}\ln\frac{2}{\{1+(2x/d)^2\}^{1/2}} + \frac{\pi}{16}\frac{w^2}{d}\frac{1}{1+(2x/d)^2}$$

where d is the separation between the electrodes, x is the distance of the earthed slit from their ends and w is the gap between the slit jaws (Hertzog 1935). The ends of the electrodes and the position of the slits were arranged so that the effective entrance and exit planes subtended an angle of $\pi 2\backslash 2$ at the centre of curvature of the plates to obtain correct focusing.

Since it was assumed that the particles described a circular path until they reached the effective exit plane and then continued tangentially until reaching the exit slit, the exact determination of the position of the exit plane relative to the exit slit was necessary. Uncertainty in this determination produced a possible error of up to $\pm 1{\cdot}5$ parts in 10^4 in the final energy measurement when the double exit slits were used, and 0·5 part in 10^4 when a fixed single slit was used.

5. *Errors caused by fringing fields due to the finite depth of the analyser plates.* The effect of the edges of the deflector bars has been calculated to produce a field inhomogeneity of one part in 10^5 over the depth occupied by the beam. This is negligible.

6. *Errors in measurement of stray magnetic fields.* The component of magnetic field between the deflector plates and perpendicular to the baseplate, was the only one effective in modifying the final energy determination. The path and energy equations for a proton in a combined electrostatic and magnetic field have been solved and indicate that if the contribution of the magnetic field is to be less than one part in 10^4 then $(Ha/\mathbf{c})(\boldsymbol{e}/2\boldsymbol{m}V)^{1/2} \leqslant 10^{-4}$ where a is the radius of curvature of the path and other symbols have their usual significance; gaussian units are used.

The magnetic field between the deflector plates was investigated, using a flip coil and sensitive galvanometer, and found to have a mean value of 0·1 gauss

out from the baseplate. This produced an error of less than one part in 10^4 for beam energies above 100 kev.

7. *Errors associated with targets.* Using the technique described in § 3 (iv), it is estimated that the surface contamination of the target had been reduced to a negligible amount. Half the energy lost by the beam in traversing the target was subtracted from the energy of the observed peaks to obtain the true resonant energy (Morrish 1949). Uncertainty in the energy loss was estimated at $\pm 10\%$, which produced an uncertainty of $\pm 3 \times 10^{-4}$ in results obtained using the thicker targets. For the 500 ev thick targets the uncertainty was less than 10^{-4}.

8. *Orthogonality of the entrance plane of the analyser with the beam.* This was checked by rotating the analyser through 180° about a vertical axis and repeating resonance determinations of the two fluorine lines (Runs 11 and 12, table 1). There was no significant difference between the two sets of measurements, which indicated that the beam was vertical. The accuracy with which the entrance plane of the analyser could be set horizontally was calculated to produce an error of $\pm 2 \times 10^{-4}$ in the final energy determination, and slight fluctuations in beam direction were calculated to produce a similar error.

9. *Uncertainties produced by the statistical accuracy of counting.* With the exceptions of Runs 1 and 2, statistical accuracy of counting was sufficiently high to enable the energy of maximum yields to be read from the experimental curves with an accuracy to $\pm 0{\cdot}1$ kev for the sharp fluorine lines, and to $\pm 0{\cdot}2$ kev for the broader lithium line.

Estimation of Overall Accuracy. Of the above errors, only Nos. 2, 3 and 4 were purely systematic; Nos. 8 and 9 were purely random, and the others had both a random and a systematic component.

The root of the squares of the purely random errors ($\pm 3 \times 10^{-4}$) agreed well with the standard deviation of individual determinations listed in table 1, which confirmed that no large random error had been overlooked. The standard deviations of the three mean values given in table 1 were between 2×10^{-4} and 4×10^{-4}.

The root of the squares of all errors which could have a systematic component was 4×10^{-4}. It is felt that an overall error of less than $\pm 1 \times 10^{-3}$ for the final mean values can therefore be claimed with some confidence.

(ii) *Errors affecting the Measurement of Half Widths*

Of the above errors, only those associated with targets and statistical accuracy affect the determination of the width of the resonance curve at half maximum yield. Estimations of half width have been made only when the energy lost in the target was small compared with the natural half widths, so that the uncertainty in the target correction given in § 3 (iv) of the paper was negligible.

The possible inhomogeneity of the beam (one part in a thousand) was too small to add significantly to the observed widths.

The dead time of the counter and quench unit was calculated to cause an error of -3% in the maximum counting rate and $1\frac{1}{2}\%$ in the counting rate at half maximum for the strongest fluorine line using a 2 kev LiF target, but in most cases the error was much less so that the broadening of the resonances due to this cause was negligible. The error associated with statistical accuracy of counting therefore remains predominant; as this varied from one determination to another the estimated errors have been quoted separately in table 1.

Neutron Total Cross Sections

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MS. received 18*th June* 1952

ABSTRACT. The neutron total cross sections of Be, Na, Mo and Hg have been measured in the energy region 1 ev to 10 kev, and of Th between 1 ev and 1 kev, using the Harwell linear accelerator time-of-flight spectrometer. The energy resolution is better than in previously reported work on these elements, and several resonances have been more clearly resolved.

Some measurements have been made, by the transmission method, of the neutron total cross sections of beryllium, sodium, molybdenum and mercury in the energy range 1 ev to 10 kev, and of thorium between 1 ev and 1 kev, using the Harwell linear accelerator time-of-flight spectrometer. The apparatus has been adequately described in other papers (Merrison and Wiblin 1951, Goulding *et al.*, paper to be published). The neutrons were timed over a flight path of 10 metres for the Be, Na, Mo and Hg measurements, but since the diameter of the Th sample was only 1 inch it was necessary to use a 4-metre flight path to achieve a reasonable counting rate. Figures 1 to 5 give total cross sections in barns (10^{-24} cm^2/atom) plotted against neutron energy in electron volts. The energy resolutions are shown at 10, 100 and 1000 ev; those for Hg applying also to Mo and Na. Wherever possible, estimates of $\sigma_0\Gamma^2$ for the resonances were obtained from the transmission curves by the area method using the Breit–Wigner one-level formula (Rainwater and Havens 1946, Havens and Rainwater 1951 a). σ_0 is the true total cross section at exact resonance, and Γ the width of the resonance curve at half-height.

The resonance cross sections quoted are those actually measured, and the accuracies indicated are those corresponding to the counting statistics. The measured resonance cross sections may be very much smaller than the true values, due to the poor resolution of the spectrometer relative to the resonance widths, and from the experimental data it is not possible to estimate σ_0 and Γ separately. Nevertheless, the values of $\sigma_0\Gamma^2$ are significant. Estimates of $\sigma_0\Gamma^2$ are best made with a thick sample ($n\sigma_0 \gg 1$, where n is the number of atoms/cm^2 of natural element) and it is theoretically possible to determine $\sigma_0\Gamma$ from measurements on a thin sample ($n\sigma_0 \ll 1$) (Rainwater and Havens 1946, Havens and Rainwater 1951 a). It was not, however, practicable to carry out such measurements involving much longer counting times (to achieve reasonable statistical accuracy) with the existing experimental conditions. These measurements should be feasible with the higher counting rates expected of the 15 Mev linear accelerator spectrometer now being installed.

Analyses of the samples have not revealed any impurities in sufficient quantities to affect the results.

* On leave from the University of Otago, New Zealand.

Beryllium (fig. 1). The sample was of metal compacted from powder, and machined to a thickness of 2·36 g/cm². The cross section is constant in the region measured, the best fit to the statistically weighted points above 5·5 ev giving 6·04 ± 0·03 barn, which agrees with the results reported by Adair (1950).

Sodium (fig. 2). Two vacuum distilled samples of thickness 9·26 g/cm² and 1·26 g/cm² were used. These samples showed a large resonance in the cross section, rising to 34·3 barn at ~3500 ev, and below the resonance a constant cross section of 2·98 ± 0·02 barn. Values of $\sigma_0\Gamma^2$ of 58×10^6 barn (ev)² for the

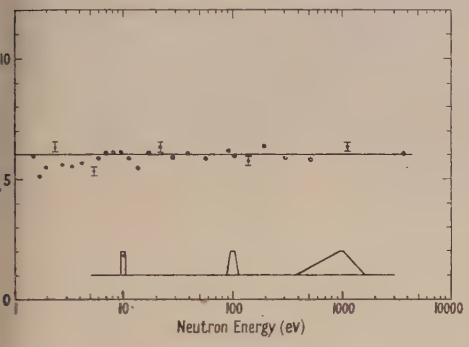

Fig. 1. The total cross section of beryllium, showing the resolution of the spectrometer.

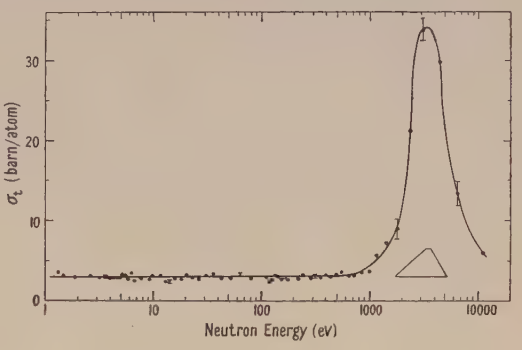

Fig. 2. The total cross section of sodium, showing the resolution of the spectrometer at the resonance energy.

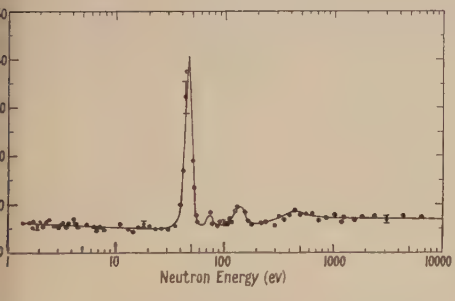

Fig. 3. The total cross section of molybdenum.

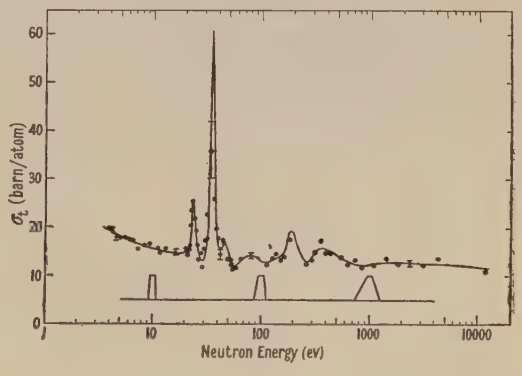

Fig. 4. The total cross section of mercury, showing the resolution of the spectrometer.

large sample, and 57×10^6 barn (ev)² for the small sample were obtained. In this region, however, the energy resolution of the spectrometer is only 1/1·1.

Molybdenum (fig. 3). Two samples of thickness 15·6 g/cm² and 3·82 g/cm² were used.

A sharp resonance in the cross section, rising to 40·2 barn, was observed at 46·3 ev, with $\sigma_0\Gamma^2 = 400$ barn (ev)². Further small resonances were observed at 75 ev, 140 ev and ~440 ev.

Accepting the results of Egelstaff and Taylor (1950) below 10 ev, the dip in cross section around 25 ev appears to be significant, indicating that the resonance at 46 ev may be due to scattering. It is hoped to investigate this more accurately with the new 15 MeV linear accelerator spectrometer.

Mercury (fig. 4). Two samples of thickness 14·22 g/cm² and 34·42 g/cm² from a supply of vacuum redistilled liquid mercury were used.

This curve is interesting in that the resonance indicated by Havens and Rainwater (1946) above 25 ev has been resolved into two levels at 23·3 and 35·4 ev, showing total cross sections of 25·3 and 60·9 barn, and $\sigma_0\Gamma^2$ of 9 and 170 barn (ev)² respectively.

Further resonances of 19·3 barn at 191 ev, and 17·0 barn at ~350 ev were also resolved.

Thorium (fig. 5). Two samples of thickness 18·75 g/cm² and 27·65 g/cm² were obtained from a bar made by cold pressing and sintering thorium powder.

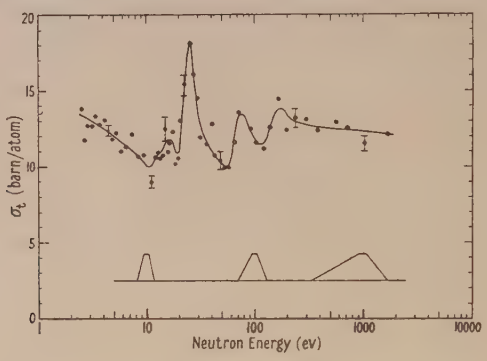

Fig. 5. The total cross section of thorium, showing the resolution of the spectrometer.

The resonances reported by Havens and Rainwater (1951 b) at 23 ev and 84 ev have been clearly resolved, but were observed at 25 ev and 80 ev, with a broader resonance at 170 ev. The shape of the curve in the region of 15 ev is probably due to several incompletely resolved resonances. The 25 ev resonance has $\sigma_0\Gamma^2 = 64$ barn (ev)².

ACKNOWLEDGMENTS

We are indebted to Mr. E. R. Wiblin, under whom the work was carried out, and to Dr. E. Bretscher and the Director of the Atomic Energy Research Establishment for permission to publish this work. We also wish to thank Mr. W. B. Woollen, Miss J. R. Murray and Mrs. B. M. Childs for the preparation and analyses of the samples.

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Nuclear Disintegrations by Slow Negative μ-Mesons and Photons

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Communicated by H. Fröhlich; *MS. received* 16*th May* 1952

ABSTRACT. The emission of particles from a nucleus following the capture of a negative μ-meson is discussed using evaporation theory. Assuming a reaction of the form $\mu^- + P \rightarrow N + \mu^0$, it is possible to calculate a distribution of nuclear excitation from which the mean probability of charged particle emission is evaluated.

In the relation between excitation U and nuclear temperature τ the exponent of τ is given the values 2 and 3. The usual law $U = \lambda\tau^2$ is found to predict results for proton emission which are far less than those experimentally observed.

In order to investigate this energy–temperature relationship data from low energy photo-disintegration experiments ($U \simeq 20$ MeV) have been compared with theoretical values. Combination of the limited evidence available with very high energy data suggests a variation of excitation with the cube of the nuclear temperature.

§ 1. INTRODUCTION

In the work of Sard and collaborators (1948, 1949, 1952) the absorption of negative μ-mesons in lead was investigated by studying the production of neutrons from the excited nucleus. Their most recent estimation of the number of neutrons per captured μ^--meson is $2{\cdot}16 \pm 0{\cdot}15$, with an additional error of $\pm 10\%$ due to the uncertainty in the strength of the standard neutron source used to determine neutron detecting efficiency.

This result is in reasonable agreement with those of Groetzinger *et al.* (1948, 1949, 1951) whose latest experiments give a value of $1{\cdot}92 \pm 0{\cdot}72$ neutrons per disintegration.

While the emission of neutrons had thus been fairly well established for some time, there seemed to be no experiments which showed conclusively the emission of charged particles after the absorption of the μ^--meson. Several attempts were made to investigate this both at mountain altitude and at sea level (Wang and Jones 1948, Chang 1949, Cool *et al.* 1949, Perkins 1949), but the results indicated that any charged particle emission was, at the best, a very rare event.

The principal difficulty encountered in these investigations is the constitution of the mesons employed, since the slow mesons of cosmic radiation which are stopped in the absorbers consist of a mixture of negative π- and μ-mesons. It is well known that the π^--mesons are extremely efficient at producing nuclear disintegrations with emission of charged particles and hence, in the experiments performed, it is not entirely clear which type of meson is involved.

By working underground (George and Evans 1951) at various depths down to about 60 m water equivalent, the ratio of π^-/μ^--mesons has been considerably reduced, and it has been possible to separate out the disintegrations due to μ^--meson capture from the few that may still be attributed to π^--mesons.

Observations were made with the aid of nuclear emulsions from which the experimental value obtained for the average number of prongs per disintegration is $0{\cdot}1 \pm 0{\cdot}02$.

It is assumed that the charged particles emitted are protons and thus, in the following, an attempt is made to calculate the possible number of protons per stopped μ^--meson using evaporation theory.

The required knowledge of the energy given to the nucleus following the absorption of a μ^--meson offers a direct link with the observed neutron emission. An estimation of this nuclear excitation has been made employing the charge-exchange reaction $\mu^- + \mathrm{P} \rightarrow \mathrm{N} + \mu^0$. It is shown that the emission of 2 to 3 neutrons from lead can be explained by modifying the calculation of Tiomno and Wheeler (1949).

The question of the relation between the excitation of the nucleus U and the nuclear temperature τ is seen to arise, and, in the first place, a general expression $U = A\tau^{\delta}/K$ is assumed, where A is the mass number and K is constant. In subsequent calculations δ is given the values 2 and 3.

The meson data themselves do not provide sufficient evidence from which to draw any definite conclusions on this energy–temperature relation. Thus, it has been necessary to search for some other source providing nuclear disintegrations in the same energy range and with which it is perhaps permissible to make a comparison.

To this end the last section is devoted to a consideration of a number of photo-disintegration results. The data are taken from the work of Hirzel and Wäffler (1947) and Mann and Halpern (1951). The former have investigated various isotopes in the range $50 \leqslant A \leqslant 118$ employing gamma-radiation of $h\nu = 17{\cdot}2$ Mev and have obtained the ratio of the cross sections $\sigma(\gamma, \mathrm{p})/\sigma(\gamma, \mathrm{n})$, while the latter have carried out experiments in a rather larger range $4 \leqslant Z \leqslant 50$ irradiating each target with a bremsstrahlung spectrum of maximum energy $23{\cdot}5$ Mev. Their results together with those of Price and Kerst (1950) on γ–n reactions have led to an evaluation of the yield ratio N/P.

Both these ratios may be compared directly with the values deduced from theory to indicate the nuclear temperature at an excitation energy of about 20 Mev. Throughout the following calculations reference is made to the investigation of disintegration stars in silver and bromine as carried out by Le Couteur (1950, referred to as LeC) and it is assumed that the formulae of LeC give a good representation of the nuclear temperature at a mean excitation of 200 Mev.

It appears that the results of this paper are best connected to this high energy data by assuming that the nuclear excitation varies as the cube of temperature.

§ 2. THE PROBABILITY OF PROTON EMISSION

An expression for the probability per unit time of the emission of a particle with kinetic energy T from a nucleus a leaving a residual nucleus b has been derived by Weisskopf (1937) as

$$P(T)\,dT = \frac{\sigma g m T}{\pi^2 \hbar^3} \frac{\rho(b)}{\rho(a)}\,dT, \qquad \ldots\ldots(2.1)$$

where ρ is the level density, g the number of spin states of particle, and m its mass.

According to Bethe (1937), the level density in terms of nuclear excitation U, temperature τ and entropy S may be written as

$$\frac{1}{\rho(U)} = \tau\left(2\pi \frac{dU}{d\tau}\right)^{1/2} \exp(-S). \qquad \ldots\ldots(2.2)$$

Thus, in order to evaluate the probability, it is necessary to assume some relation between the excitation U and temperature τ.

In the following paragraphs, the variation of U with both τ^2 and τ^3 is considered and so it is convenient at this point to use a generalized form

$$U = A\tau^\delta/K \qquad \ldots\ldots(2.3)$$

where A is the mass number and K a numerical constant for a particular δ. For the purposes of the immediate calculation let $A/K=\lambda$. By consideration of LeC, eqns. (52) and (53), (2.1) becomes

$$P(T)\,dT = \gamma(T-V')\left(\frac{U_a}{U_b}\right)^{(\delta+1)/2\delta} \exp(S_b - S_a)\,dT, \qquad \ldots\ldots(2.4)$$

where V' is the effective height of the potential barrier for the particle and $\gamma=\sigma_0 gm/\pi^2\hbar^3$ as defined by LeC (51). Using the notation $T=U_a-Q-U_b$, $R=U_a-Q-V'$, Q being the nucleon binding energy in the parent nucleus, it is possible to determine the total emission probability per unit time, and mean energy of emission from

$$\int_{V'}^{U_a-Q} P(T)(T-V')^n\,dT = \gamma\int_{V'}^{U_a-Q} (T-V')^{n+1}\left(\frac{U_a}{U_b}\right)^{(\delta+1)/2\delta} \exp(S_b-S_a)\,dT. \qquad \ldots\ldots(2.5)$$

To a first approximation the probability is

$$P(T)\,dT = \gamma\left(\frac{U_a}{R}\right)^{(\delta+1)/2\delta} \tau_R{}^2 \exp(S_R - S_a), \qquad \ldots\ldots(2.6)$$

and the mean energy of emission may be written simply as $T=V'+2\tau_R$.

Relative probabilities of particle emission may be determined from (2.6). Since only low excitation energies U_a less than 40 Mev are being employed, it is necessary merely to consider the competition between neutrons and protons, and the expression becomes

$$\frac{P_n}{P_p} = \frac{\gamma_n}{\gamma_p}\left(\frac{\lambda_p}{\lambda_n}\right)^{2/\delta}\left(\frac{R_n}{R_p}\right)^{(3-\delta)/2\delta} \exp(S_{R_n} - S_{R_p})$$

$$= \frac{\gamma_n}{\gamma_p}\left(\frac{\lambda_p}{\lambda_n}\right)^{2/\delta}\left(\frac{R_n}{R_p}\right)^{(3-\delta)/2\delta} \exp\frac{\delta}{\delta-1}(\lambda_n{}^{1/\delta}R_n{}^{(\delta-1)/\delta} - \lambda_p{}^{1/\delta}R_p{}^{(\delta-1)/\delta}), \qquad \ldots\ldots(2.7)$$

where R_x is the excitation of the nucleus b after emission of a particle x(n, p) from the initial state a, and $\gamma_n=\gamma_p$ (LeC (59)).

For $\delta=2$ the term $(\lambda_p/\lambda_n)(R_n/R_p)^{1/4}$ outside the exponential may be replaced by unity with negligible error. This approximation is even better for $\delta=3$, when the exponent of R_n/R_p becomes zero and a lower power of λ_p/λ_n is involved.

Hence, the effective probability ratio, in these cases, is

$$\frac{P_n}{P_p} = \exp(S_{R_n} - S_{R_p}) = \exp\left\{\frac{\lambda^{1/\delta}(R_n - R_p)}{R_{np}{}^{1/\delta}}\right\} = \exp\left\{\frac{\lambda^{1/\delta}(Q_p - Q_n + V_p')}{R_{np}{}^{1/\delta}}\right\}, \qquad \ldots\ldots(2.8)$$

where

$$2R_{np}{}^{1/\delta} = R_n{}^{1/\delta} + R_p{}^{1/\delta}. \qquad \ldots\ldots(2.9)$$

With $\delta=2$ and using LeC (37), it is found that

$$\frac{P_n}{P_p}=\exp\left\{\frac{\Lambda(Q_p-Q_n+V_p')}{2(R_{np})^{1/2}}\right\}. \qquad \ldots\ldots(2.10)$$

The value chosen for the nuclear parameter Λ is $(A/3{\cdot}3)^{1/2}$ (LeC § 4.1).

Consider a nucleus (A, Z) of binding energy B_Z^A. Then $Q_p=B_Z^A-B_{Z-1}^{A-1}$, $Q_n=B_Z^A-B_Z^{A-1}$ or $Q_p-Q_n=B_Z^{A-1}-B_{Z-1}^{A-1}$. This, in turn, may be written in terms of the isobaric energy difference $Q_p-Q_n=E_{Z-1}^{A-1}-E_Z^{A-1}-(m_n-m_p)$, m_n, m_p being mass of neutron and proton respectively. Q_p-Q_n may then be evaluated from a knowledge of the $\beta^{\pm}$-energies of the residual nuclei.

(i) *Proton Emission at the Second Stage*

The mean reduction in energy due to the emission of a particle x(n, p) is

$$H_x=Q_x+V_x'+2(R_x/\lambda)^{1/\delta}, \qquad \ldots\ldots(2.11)$$

so arriving at a value for the remaining excitation which will be denoted by U_a'.

Owing to the effects of V_x', a far greater probability of proton emission at this second stage exists if the first particle x is a neutron. Because of this, x = n is used throughout the calculations of H_x. There is an extremely small probability of protons being emitted at both first and second stages, but only at the upper limit of the energy range employed. The consequent reduction in excitation for a nucleus at the third stage is such as to make further proton emission most improbable in the present case.

It has been assumed, therefore, that the total probability of proton emission per disintegration may be represented by the sum $P_p+P_p'=(P_p)_T$. Since only neutrons and protons are considered to be emitted, it is simple to deduce P_p as the absolute proton emission probability at the first stage and P_p' as the absolute probability at the second.

(ii) *Correction to the Probability P_p'*

It will be seen that in the evaluation of H_x (2.11) the energy of the emitted particle x is assumed to have its mean value. In point of fact the residual energy U_a' is dependent upon the energy spectrum of the emitted particle and consequently calculations have been carried out to examine the extent of such dependence.

Let the distribution of particle energy be represented by the function $\rho(U_a')$. Then the true expression for P_p' becomes

$$P_p'=\int\rho(U_a')P_p'(U_a')\,dU_a'. \qquad \ldots\ldots(2.12)$$

By expansion this is

$$\int\rho(U_a')[P_p'(\overline{U_a'})+(U_a'-\overline{U_a'})(P_p')'+\tfrac{1}{2}(U_a'-\overline{U_a'})^2(P_p')''\ldots.]\,dU_a',$$

which on integration gives

$$P_p'=P_p'(\overline{U_a'})+\tfrac{1}{2}\overline{(U_a'-\overline{U_a'})^2}\,\frac{d^2P_p'}{dU_a'^2}. \qquad \ldots\ldots(2.13)$$

From (2.5), $\overline{(U_a'-\overline{U_a'})^2}$ may be expressed in terms of the nuclear temperature as $\overline{(U_a'-\overline{U_a'})^2}=2\tau^2$. Hence, the corrected expression for the probability P_p' becomes

$$P_p'=P_p'(\overline{U_a'})+\tau^2\,\frac{d^2P_p'}{dU_a'^2}. \qquad \ldots\ldots(2.14)$$

It appears from fig. 4 that the correction is extremely small.

The calculation of $(P_p)_T$ involves both the effective potential barrier and the nuclear parameter Λ. If the barrier height V is evaluated from the expression of Bethe (1937) it is then in reasonable accordance with the penetration formula of Bethe and Konopinski (1938) to assume the relationship $V_p' = 0{\cdot}7V$. The bromine isotope $A = 79$, $Z = 35$, thus gives an effective potential barrier $V_p' = 4$ MeV. In LeC §4.1, case *d*, the parameter Λ is given the value $\sqrt{(A/3{\cdot}3)}$ but in the following the proton probability has been calculated for Λ ranging from $\sqrt{(A/3{\cdot}3)}$ to $0{\cdot}86\sqrt{(A/3{\cdot}3)}$. It is not considered possible to reduce Λ further and still obtain results in fair agreement with the disintegration star data upon which the numerical value of Λ is based (LeC).

§3. PROBABILITY DISTRIBUTION OF NUCLEAR EXCITATION ENERGY AND MEAN EMISSION PROBABILITY

To apply the emission probabilities to the problem of μ^--meson capture a knowledge of the probability of a given nuclear excitation energy and of its maximum possible value due to the capture process is necessary. It is essential to be able to link up theoretical predictions with experimental fact, for, in order that any progress may be made, the value of the maximum energy available for the nucleus must be fairly well known. Fortunately, evidence can be obtained from the investigations on neutron emission due to μ^--capture. The experiments of Groetzinger, Berger and McClure (1951), and, most recently, Crouch and Sard (1952), indicate that the number of neutrons emitted per disintegration, from a lead target, lies in the range of 2 to 3. Consequent calculations show that the emission of three neutrons from a lead nucleus is only possible with an initial excitation U_a greater than 28 MeV for $\delta = 2$, eqn. (2.3), or U_a greater than 31 MeV for $\delta = 3$. If it is to be assumed, therefore, that an evaporation process occurs then it must be taken that the excitation energy of the nucleus due to a stopped μ^--meson has a maximum value considerably in excess of that deduced in previous calculations by Tiomno and Wheeler (1949).

Tiomno and Wheeler calculated the energy given to the nucleus by μ^--capture assuming a reaction of the form $\mu^- + \mathrm{P} \rightarrow \mathrm{N} + \mu^0$. Here the nuclear excitation is due to the recoil momentum of the neutron, and the effects are considered using various nuclear models.

For the Fermi gas model the excitation is estimated to have a maximum value at approximately 22 MeV, this being calculated on the assumption that μ^0 has zero mass and $\mu = 210\,m$. They obtain a result for the distribution of nuclear excitation which is proportional to

$$I\left(\frac{Q}{mc^2}\right) = m^{-3}c^{-4}(\mu c^2 - Q)2MQ \qquad \text{for} \qquad 0 \leqslant Q \leqslant Q^*$$

$$= m^{-3}c^{-4}(\mu c^2 - Q)\left[P^2 - \left(\frac{MQ}{P_0} - \frac{P_0}{2}\right)^2\right] \qquad \text{for} \qquad Q^* \leqslant Q \leqslant Q_{\max}. \qquad \ldots\ldots (3.1)$$

Q and P measure the excitation energy and maximum Fermi momentum respectively.

If, however, the free particle model is corrected according to Bardeen (1937), the excitation of the nucleus may be increased quite considerably. The modification introduced by using Bardeen's correction results in a reduction in the effective mass of the nucleon to $M_{\mathrm{eff}} = M/\gamma$ where γ is the Bardeen number.

It is thus possible to replot the curves of Tiomno and Wheeler substituting the reduced mass M_{eff} in (3.1). Two distributions are given in fig. 1, where X and Y correspond to values of the Bardeen number $\gamma=1{\cdot}5$ and $\gamma=2$, respectively. The original curve W is also given to facilitate comparison. Values of γ in the range $1\leqslant\gamma\leqslant2$ are considered, these being in reasonable agreement with the values suggested from the disintegration star evidence (LeC).

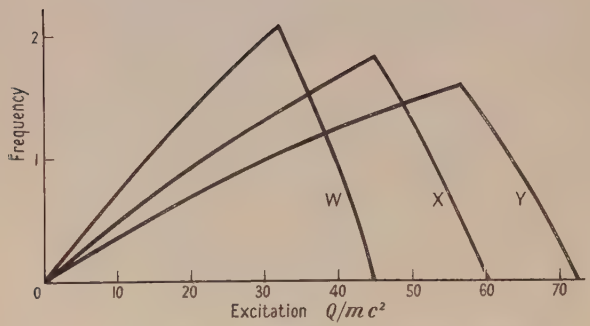

Fig. 1. Frequency distribution of excitation energy in the nucleus due to the capture of a slow negative μ-meson assuming the reaction $\mu^-+\text{P}\rightarrow\text{N}+\mu^0$. W: Fermi model (Tiomno and Wheeler 1949). X, Y: with Bardeen's correction ($\gamma=1{\cdot}5$ and 2 respectively).

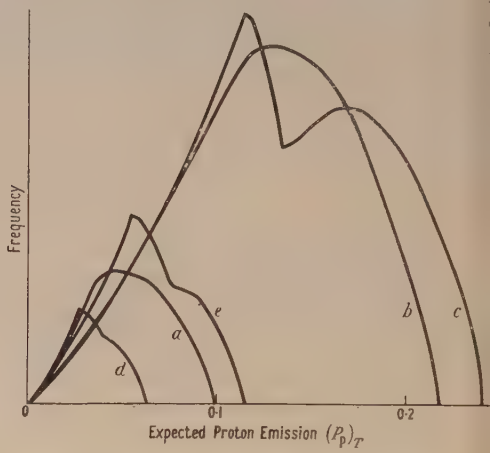

Fig. 2. Possible frequency distributions of expected total proton emission from various nuclei assuming a nuclear excitation distribution as given in fig. 1.
a: ^{79}Br, $\delta=2$, $\gamma=1{\cdot}5$. *c*: ^{79}Br, $\delta=3$, $\gamma=1{\cdot}5$.
b: ^{79}Br, $\delta=2$, $\gamma=2$. *d*: ^{81}Br, $\delta=3$, $\gamma=1{\cdot}5$.
e: ^{109}Ag, $\delta=3$, $\gamma=1{\cdot}5$.

By integration of $(P_p)_T$ (as given in figs. 3 and 5) over the corresponding distribution in nuclear excitation, an attempt has been made to predict the mean probability of proton emission per disintegration in the capture process. Curves showing the possible distribution of $(P_p)_T$ for various values of δ and γ are given in fig. 2.

§4. NUMERICAL VALUES FOR μ^--INDUCED DISINTEGRATIONS

(i) *Using a Square Law* $\delta=2$

With $\delta=2$, evaporation theory assumes its more common form and, from (2.10) together with the value $\Lambda=\sqrt{(A/3{\cdot}3)}$ and its lower limit, it has been possible to calculate the expected probability $(P_p)_T$ for various initial excitation energies. Graphical representation is given for isotopes of bromine and silver in fig. 3.

The correction to P_p' (2.14) was calculated for the bromine isotope $A=79$ as shown in fig. 4. The result is a small positive increase in the probability which becomes of negligible effect as the excitation energy is increased. Further, on considering the total mean probability over the available energy range as indicated in §3, the contribution of the correction was of such small account that it was considered possible to neglect the effect in further calculations.

The problem of allocating an appropriate Bardeen number to a particular value of the parameter Λ is simply solved by reference to the relation LeC (39). Here, approximately, $\Lambda^2=\pi^2A/\zeta\gamma$ where $\zeta=22$ Mev. It is then seen that a

reduction of Λ by 14% corresponds to an increase in γ to the approximate value 2. Therefore, the sets of parameters employed are $\Lambda=\sqrt{(A/3{\cdot}3)}$, $\gamma=1{\cdot}5$; $\Lambda=0{\cdot}86\sqrt{(A/3{\cdot}3)}$, $\gamma=2$. The probabilities of proton emission are presented

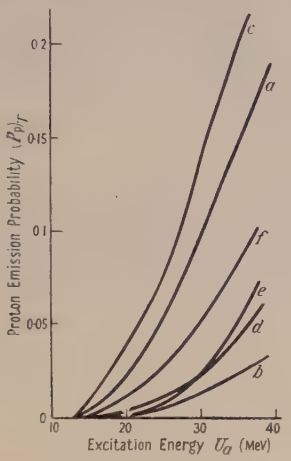

Fig. 3. Estimation of the total proton emission probability $(P_p)_T$ for various isotopes when $\delta=2$. *a*, *c*: ^{79}Br; *b*, *d*: ^{81}Br; *e*: ^{107}Ag; *f*: ^{109}Ag. $\Lambda=\sqrt{(A/3{\cdot}3)}$ for curves *a*, *b*, $\Lambda=0{\cdot}86\sqrt{(A/3{\cdot}3)}$ for curves *c*–*f*.

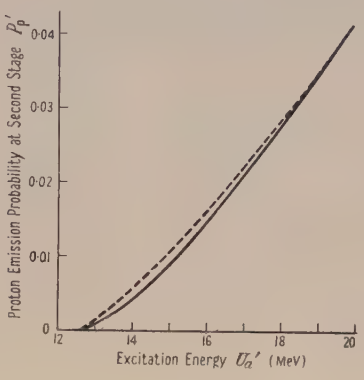

Fig. 4. The solid curve is the proton emission probability P_p' at the second stage assuming that a neutron of mean energy $H_n=Q_n+4R_n^{1/2}/\Lambda$ has been emitted at the first stage. The dotted curve indicates the corrected P_p' taking into account the energy spectrum of the emitted neutron.

in table 1. It will be seen that the contributions from the silver isotopes have been calculated merely for the reduced Λ, as it was found that only in this case were they appreciable compared with those from bromine.

From the prong distribution of George and Evans (1951) the proton number per μ^--meson induced disintegration is $0{\cdot}1\pm0{\cdot}02$, a value which is not reached by even the upper limit of the theoretical predictions. It should be remembered

Table 1

$\Lambda=\sqrt{(A/3{\cdot}3)}$, $\gamma=1{\cdot}5$			$\Lambda=0{\cdot}86\sqrt{(A/3{\cdot}3)}$, $\gamma=2$		
Isotope	$(P_p)_T$	Mean prob. for AgBr	Isotope	$(P_p)_T$	Mean prob. for AgBr
^{79}Br	0·053		^{79}Br	0·125	
^{81}Br	0·006	~0·015	^{81}Br	0·022	0·058
			^{107}Ag	0·032	
			^{109}Ag	0·052	

that this higher theoretical result has only been reached by employing rather extreme values for the parameters Λ and γ. It would be more satisfactory if the unmodified Λ and lower γ gave reasonable agreement with experiment.

On the other hand, it is possible that some direct 'knock-out' process occurs with a frequency sufficient to increase suitably the proton yield. Experimentally it is not yet clear whether there is any evidence of the occurrence of this type of process in the μ -meson capture, but experiments on photo-disintegration seem to indicate that the effect of any direct interaction between quantum and proton is small compared with that from compound nucleus formation (§ 5).

Lack of evidence in the meson-capture experiments makes it difficult to say whether it is possible to draw an analogy between the two cases, but it was thought worthwhile to search for some other source from which increased proton emission might be obtained. To this end, investigations were carried out to examine the effect upon evaporation theory when the excitation energy of the nucleus was assumed proportional to the cube of the nuclear temperature, and it will be seen in the following paragraphs that this treatment predicts results in much closer agreement with the experimental data.

(ii) *Using a Cube Law* $\delta = 3$

From disintegration star evidence (LeC) it seems possible to assume that the 'square' relation LeC (37), i.e. $U = \Lambda^2\tau^2/4$ is accurate for a mean excitation value $U_a = 200$ MeV in a nucleus $A = 80$. Hence, the required (U, τ) relation when $\delta = 3$ becomes

$$U = A\tau^3/75{\cdot}6. \qquad \ldots\ldots(4.1)$$

It is obvious that this expression is still dependent upon the nuclear parameter Λ and has, in fact, been deduced from the value $\Lambda = \sqrt{(A/3{\cdot}3)}$. So that the following results may the more easily be compared with the preceding set, the

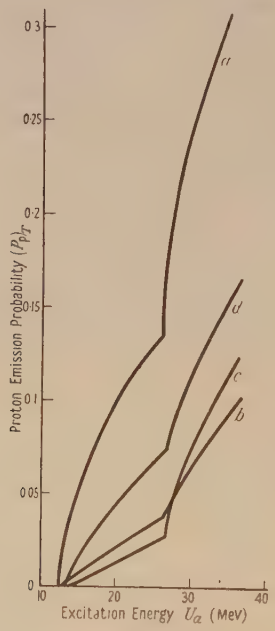

Fig. 5. Estimation of the total proton emission probability $(P_p)_T$ for $\delta = 3$, $\Lambda = \sqrt{(A/3{\cdot}3)}$. a: ^{79}Br; b: ^{81}Br; c: ^{107}Ag; d: ^{109}Ag.

proton probabilities have been calculated using (4.1) and also with a further similar relation corresponding to the reduced $\Lambda = 0{\cdot}86\sqrt{(A/3{\cdot}3)}$. The ratio of neutron and proton probabilities, when $\delta = 3$, is given from (2.8) and (2.9) as

$$\frac{P_n}{P_p} = \exp\{(A/75{\cdot}6)^{1/3}(Q_p - Q_n + V_p')/R_{np}^{1/3}\}. \qquad \ldots\ldots(4.2)$$

The variation of the total absolute proton probability with increasing excitation is much more rapid than in the preceding section, as illustrated in

fig. 5. Considering emission of protons at the second stage, there is, as before, a small correction (neglected in the actual calculations) for the neutron energy spectrum. The correction differs from the previous one in that it is negative in sign for energies $U_a \geqslant 27$ MeV.

Table 2

$\Lambda=\sqrt{(A/3{\cdot}3)}$, $\gamma=1{\cdot}5$			$\Lambda=0{\cdot}86\sqrt{(A/3{\cdot}3)}$, $\gamma=2$		
Isotope	$(P_p)_T$	Mean prob. for AgBr	Isotope	$(P_p)_T$	Mean prob. for AgBr
^{79}Br	0·135		^{79}Br	0·238	
^{81}Br	0·029	0·066	^{81}Br	0·074	0·137
^{107}Ag	0·037		^{107}Ag	0·102	
^{109}Ag	0·061		^{109}Ag	0·135	

Numerical values for $(P_p)_T$ are given in table 2. So increased is the probability $(P_p)_T$ when $\delta=3$ that the contributions from the silver isotopes are no longer negligible, even in the less favourable case of $\gamma=1{\cdot}5$.

§ 5. PHOTO-DISINTEGRATION OF NUCLEI

Owing to insufficient evidence in the experiments of George and Evans (1951) it is impossible to draw any definite conclusions on the relation between nuclear excitation and temperature. In this connection, values have been taken from a quantity of photo-disintegration data, in which the relative cross sections $\sigma(\gamma, p)/\sigma(\gamma, n)$, or yields, have been experimentally determined.

In the work of Hirzel and Wäffler (1947) various isotopes in the range $50 \leqslant A \leqslant 118$ were subjected to bombardment by gamma-radiation of $h\nu = 17{\cdot}2$ MeV. The experimental results for the ratio of cross sections obtained were compared with the values deduced from the statistical theory (Weisskopf and Ewing 1940). Calculations using (2.10) gave results in close agreement with the latter, i.e. theoretical values for $\sigma(\gamma, p)/\sigma(\gamma, n)$ are a factor 10 to 100 less than those observed. However, application of (4.2) predicts results which are of the same order as experiment.

Similar results were found on consideration of the data of Mann and Halpern (1951). Their experiments consisted of the use of scintillation counters in the study of the yields of photo-protons from 20 elements, $A \leqslant 50$, irradiated with a bremsstrahlung spectrum having a maximum energy of 23·5 MeV. By comparison with the results of Price and Kerst (1950) on γ–n yields, it has been possible to determine the ratio of proton/neutron yields. The results show the same form as previously, the number of protons detected being greatly in excess of those predicted by (2.10). The results from the two sets of data are represented graphically for ease of interpretation (fig. 6). For $\delta=2$, the parameters $\Lambda = \sqrt{(A/3{\cdot}3)}$ and $0{\cdot}86\sqrt{(A/3{\cdot}3)}$ have been used in an attempt to show the disparity between the extreme values for $\delta=2$ and those obtained by using an unmodified parameter when $\delta=3$.

Attention must, however, be drawn at this point to other suggested mechanisms of the photo-disintegration effect. In particular, Courant (1951) attempts an explanation of the large (γ, p) cross sections found in moderately heavy nuclei in terms of a direct quantum–nucleon interaction, thus eliminating the formation

of a compound nucleus. The cross section for such an event is small compared with the probability of such an intermediate state, but it is pointed out that it need not be very great to account for experimental results. However, Courant's results are consistently lower than the data provided. Also, investigation of the angular distribution of the emitted protons gives results which do not conform entirely with that expected on the basis of a direct 'knock-out' theory.

Thus Diven and Almy (1950) found with 21 Mev x-rays on silver that the energy and angular distributions agree quite well with the statistical theory except at the upper energy limit, while results of γ–p reactions for bromine and silver by Wäffler (private communication) lead him to the conclusion that any direct quantum–proton interaction probably contributes very little to the cross section compared with a compound nucleus effect. These views are supported by investigations of the photo-disintegration of copper (Byerly and Stephens 1951) which indicate that the reaction consists mainly of an evaporation of nuclear

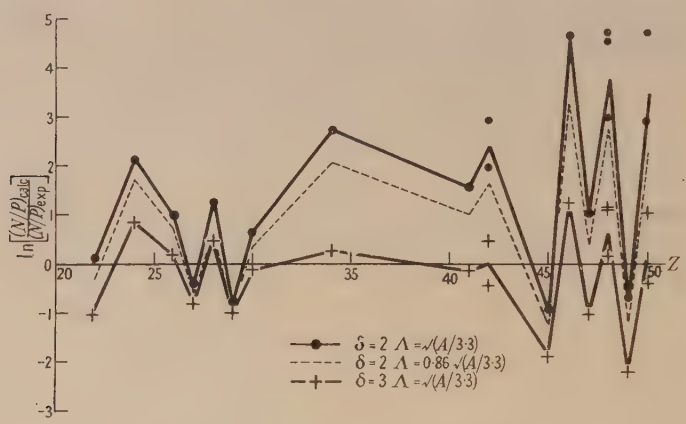

Fig. 6. Comparison of the theoretical values for relative neutron/proton yields with the data from photo-disintegration experiments.

particles. Only about 10% of the protons have energies greater than expected from evaporation, the observed angular anisotropy of these particles lending support to the suggestion of a direct photoelectric effect.

Hence, it may perhaps be assumed that any direct interaction in this range of nuclei is more or less negligible compared with an intermediate state formation, and, thus, it is necessary to turn to that form of the statistical theory in closest agreement with experiment.

Despite fairly large fluctuations about the zero point (fig. 6), it seems evident that the quantity $\ln[(N/P)_{calc}/(N/P)_{exp}]$ calculated on the basis of a cube relation between excitation and temperature comes nearest to the experimental values. Natural logarithms have been used in order that the variation of the ratio may be interpreted in terms of the difference between calculated and experimental values for the entropy $S_{R_n} - S_{R_p}$ (eqn. (2.8)).

Further, it should be pointed out that these fluctuations in results may be due to the uncertainty in the binding energies $Q_p - Q_n$. These values have been deduced from a knowledge of the $\beta^{\pm}$-activities of the nuclei after emission of the particle x(n, p), but the residual nuclei are not always well known and approximations have had to be made in several instances.

§6. CONCLUSIONS

In view of the results of the preceding section it should be permissible to suggest that the proton emission probabilities, calculated with a cube relation $\delta = 3$ and under the assumption that the formulae of LeC represent well the nuclear temperature at an excitation of 200 Mev, are the more correct. Thus it follows that, in nuclear disintegrations due to the capture of slow negative μ-mesons in silver bromide, the number of protons per disintegration is predicted as 0·066. This result may be said to be in reasonable agreement with observation, particularly when the statistical limitations of the experiment itself are taken into consideration.

It should be added that much may be learned on the subject of nuclear thermodynamics by consideration of the types of photo-disintegration experiment discussed in §5. It is necessary, however, not only that the investigations be increased in number but also that there be a considerable reduction in experimental error, before data are obtained from which reliable conclusions may be drawn.

ACKNOWLEDGMENT

The author would like to thank Dr. K. J. Le Couteur for many valuable suggestions and for his continued interest in this work.

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The Mass Difference of Sulphur 32 and Chlorine 35*

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Communicated by S. Devons; MS. received 3rd June 1952

ABSTRACT. The Q-values of the reactions $^{32}S(\alpha, p)^{35}Cl$ and $^{35}Cl(n, \alpha)^{32}P$ were measured as $-2{\cdot}02 \pm 0{\cdot}11$ MeV and $+1{\cdot}07 \pm 0{\cdot}15$ MeV respectively. From these Q-values and from the β-decay energy of ^{32}P the difference in mass excess of ^{32}S and ^{35}Cl was calculated in two independent ways, giving $1{\cdot}96 \pm 0{\cdot}11$ MeV and $1{\cdot}98 \pm 0{\cdot}15$ MeV.

In the investigation of the (α, p) reaction the nuclear emulsion technique was used in conjunction with α-particles from ThC′. An ionization chamber was used to measure the α-particle energy in the case of the (n, α) reaction which was excited by fast neutrons obtained from the D–D process.

§ 1. INTRODUCTION

THE mass difference of two nuclei can be determined either from the Q-values of nuclear reactions or from mass spectroscopic data.

For (α, p) reactions represented by the expression

$$^{A}Z(\alpha, p)^{(A+3)}(Z+1)$$

the difference in mass excess of the target and the product nuclei is

$$\Delta = (\Delta M)_Z - (\Delta M)_{(Z+1)} = Q - [(\Delta M)_{He} - (\Delta M)_H] = Q + 3{\cdot}98 \text{ MeV},$$

where ΔM is the mass excess of an isotope and Q is the reaction energy corresponding to a transition to the ground state. Similarly, for (n, α) reactions

$$\Delta = (\Delta M)_Z - (\Delta M)_{(Z-2)} = Q - 4{\cdot}76 \text{ MeV},$$

and in the case of β^- decay Δ is equal to the β-decay energy. The figures used above for the mass excesses of hydrogen, helium and the neutron are those given by Tollestrup, Fowler and Lauritsen (1950).

At the time when the experiments described in the present paper were undertaken (1949) the following information was available concerning the mass difference of ^{32}S and ^{35}Cl (fig. 1):

(*a*) The masses of ^{32}S and ^{35}Cl could be deduced from Aston's mass spectrographic measurements. The values obtained were $31{\cdot}9823 \pm (3 \times 10^{-4})$ M.U. and $34{\cdot}9811 \pm (7 \times 10^{-4})$ M.U. respectively (Mattauch and Flammersfeld 1949). The corresponding difference in mass excess was $\Delta = 1{\cdot}1 \pm 0{\cdot}7$ MeV.

(*b*) Similarly the two mass values were calculated as $31{\cdot}98089 \pm (0{\cdot}7 \times 10^{-4})$ M.U. and $34{\cdot}97877 \pm (1{\cdot}8 \times 10^{-4})$ M.U. on the basis of the measurements of Okuda and Ogata (Mattauch and Flammersfeld 1949). These corresponded to

$$\Delta = 1{\cdot}97 \pm 0{\cdot}18 \text{ MeV}.$$

(*c*) Ewald (Mattauch, private communication) measured the mass of ^{32}S and obtained $\Delta M = -17{\cdot}724 \pm 0{\cdot}018$ mM.U., in substantial agreement with Aston.†

(*d*) Haxel (1935) investigated the reaction $^{32}S(\alpha, p)^{35}Cl$ with the use of ThC′ α-particles and a proportional counter as detector. He obtained a

* This paper is an abridged version of a thesis submitted to the University of London for the degree of Ph.D.

† Ewald published a value of $-17{\cdot}728 \pm 0{\cdot}019$ mM.U. in 1951.

Q-value of $-2{\cdot}3$ MeV for the transition to the ground state. The recalculated Q-value of $-2{\cdot}1$ MeV (Livingston and Bethe 1937) led to $\Delta = 1{\cdot}9$ MeV for the difference in mass excess.

(*e*) The reaction $^{35}Cl(n, \alpha)^{32}P$ and the β-decay of ^{32}P to ^{32}S provided a further means of comparison. The energy of the β-decay, $1{\cdot}712 \pm 0{\cdot}008$ MeV (Mattauch and Flammersfeld 1949) was very well known and the Q-value of the reaction $^{35}Cl(n, \alpha)^{32}P$ was measured by Metzger, Huber and Alder (1947). They obtained $Q = +0{\cdot}44 \pm 0{\cdot}20$ MeV. This, in conjunction with the β-energy,

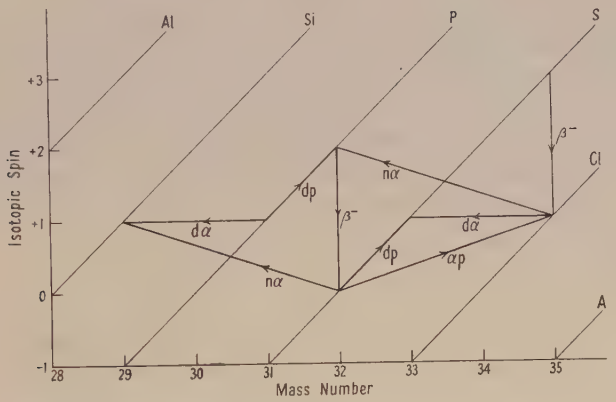

Fig. 1. The diagram of nuclei in the region of mass 29–35.

gave $\Delta = 2{\cdot}58 \pm 0{\cdot}2$ MeV. Metzger *et al.* observed that the Q-value was smaller than expected on the basis of the mass values accepted in the 1943 Isotopic Report of Fluegge and Mattauch (1943). They could not, however, suggest any reason why they should have failed to observe a more energetic α-particle group, had it been present.

(*f*) A further possible link between the two masses was provided by the reactions $^{32}S(d, p)^{33}S$ and $^{35}Cl(d, \alpha)^{33}S$. The first of these was investigated by Davison (1949), who obtained a Q-value of $6{\cdot}48 \pm 0{\cdot}11$ MeV. No reliable information existed on the second reaction, although Schrader and Pollard had reported (1941) a possible Q-value of 9·1 MeV.

The various values of Δ, the difference in mass excess of ^{32}S and ^{35}Cl, are listed below :

Reaction	Authors	Δ (MeV)
Mass spectroscopy	Aston	$1{\cdot}1 \pm 0{\cdot}7$
Mass spectroscopy	Okuda *et al.*	$1{\cdot}97 \pm 0{\cdot}18$
$^{32}S(\alpha, p)\ ^{35}Cl$	Haxel	1·9
$^{35}Cl(n, \alpha)\ ^{32}P$ and $^{32}P(\beta^-)^{32}S$	Metzger *et al.* Siegbahn	$2{\cdot}58 \pm 0{\cdot}2$
$^{32}S(d, p)^{33}S$ and $^{35}Cl(d, \alpha)^{33}S$	Davison Schrader *et al.*	1·4

In view of the lack of agreement, apparent in the table, it was decided to repeat the investigation of the reaction $^{32}S(\alpha, p)^{35}Cl$. This yielded a result in substantial agreement with that of Haxel. Then the Q-value of the reaction $^{35}Cl(n, \alpha)^{32}P$ was measured. This gave a new result. Accordingly, in what follows, the main stress is laid on the latter experiment while the former is treated very briefly.

§ 2. THE REACTION $^{32}S(\alpha, p)^{35}Cl$

A thick sulphur target was bombarded with the 8·78 MeV α-particles from ThC′. Protons emitted from the target at angles of about 90° were selected by means of a mica window, and were stopped in a nuclear emulsion. From the measurement of lengths of tracks in the emulsion the proton energies were deduced. The Q-value of the reaction was calculated according to the relation

$$E_2^0 = \frac{M_3}{M} Q + \frac{M_3 - M_1}{M} E_1,$$

where E_2^0 is the energy of the proton emitted at an angle of 90°, Q is the Q-value of the reaction, E_1 is the energy of the α-particle and M, M_1, M_3 are the mass numbers of the compound nucleus, the α-particle and the product nucleus respectively. The experimental arrangement consisted essentially of a chamber divided into two vacuum-tight and light-tight compartments. One was evacuated and contained the α-particle source and the target, while the other was air-filled and contained the photographic plates (Kodak, type NT2a). The

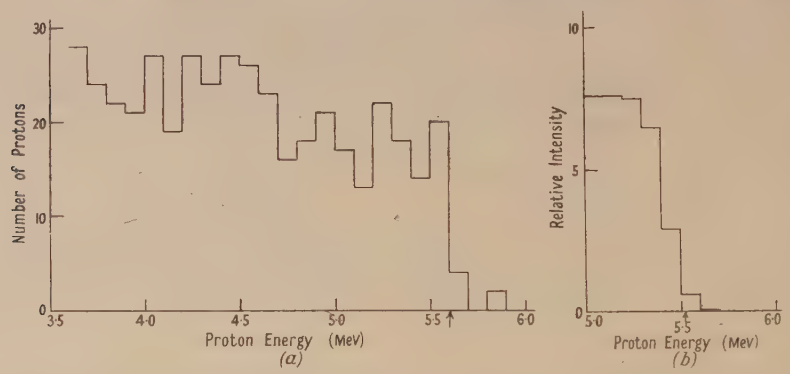

Fig. 2. (*a*) The experimental proton energy distribution for the reaction $^{32}S(\alpha, p)^{35}Cl$. (*b*) The calculated energy distribution near the end-point. The arrows indicate the values of E_2^0 corresponding to $Q = -2{\cdot}02$ MeV and $Q = -2{\cdot}1$ MeV respectively.

plates were placed so as to face the mica window situated in the partition dividing the two compartments. Screening material, the G.E.C. heavy alloy and lead, was placed wherever possible to reduce the fogging of the emulsion by γ-rays from Th C″.

The proton energy distribution obtained is shown in fig. 2(*a*). Figure 2(*b*) shows the expected shape of the distribution near the end-point, calculated on the basis of the geometry and the measured α-particle spectrum. A Q-value of $-2{\cdot}1$ MeV was assumed in the calculation, and straggling was allowed for. A comparison of the two distributions led to an experimental Q-value of $-2{\cdot}02 \pm 0{\cdot}11$ MeV. This essentially confirmed Haxel's value of $-2{\cdot}1$ MeV.

§ 3. THE REACTION $^{35}Cl(n, \alpha)^{32}P$

(i) *General*

An ionization chamber filled with carbon tetrachloride vapour was bombarded with fast neutrons. The electrical pulses from the chamber were amplified in a linear amplifier and were displayed on a cathode-ray oscillograph screen. They were photographed on a slowly moving film.

Neutrons can excite a number of reactions in the two stable isotopes of chlorine. Only two of them, however, namely $^{35}Cl(n,\alpha)^{32}P$ and $^{35}Cl(n,p)^{35}S$ have positive Q-values. In a gaseous target ionization chamber both the ejected particle and the recoil nucleus contribute to the same pulse. Accordingly all registered pulses corresponding to energies greater than the maximum neutron energy ($E_n = 3{\cdot}34$ MeV) were due to protons and α-particles from the two reactions. The geometry of the ionization chamber was such that the maximum residual energy which a proton could lose within the sensitive volume was about 1·5 MeV. Accordingly all pulses representing energies greater than that value were due to α-particles from the reaction $^{35}Cl(n,\alpha)^{32}P$.

The entire experimental arrangement was calibrated with α-particles of known energy. Thus a relation was obtained between the energy and the registered pulse height. The Q-value of the reaction was deduced from the energy E of the end-point of the pulse-height distribution according to the relation $Q = E - E_n$.

(ii) *Experimental Arrangement*

The neutron flux was obtained from the D–D reaction. A deuteron beam from a Van de Graaff generator excited the reaction in a heavy orthophosphoric acid target. The deuteron energy was 370 kev, corresponding to a maximum neutron energy of 3·34 MeV. Neutrons emitted from the target in the forward direction were used. The neutron energy spectrum was measured by means of

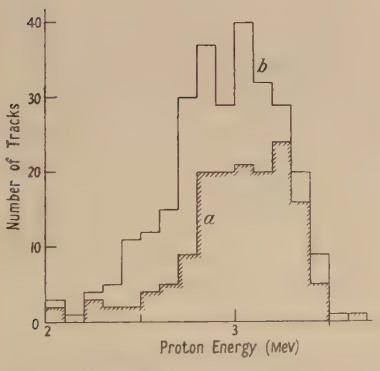

Fig. 3. The energy spectrum of recoil protons. Curve *a* includes tracks whose directions were within 10° of the forward neutron direction; curve *b* includes tracks deviating up to 30°.

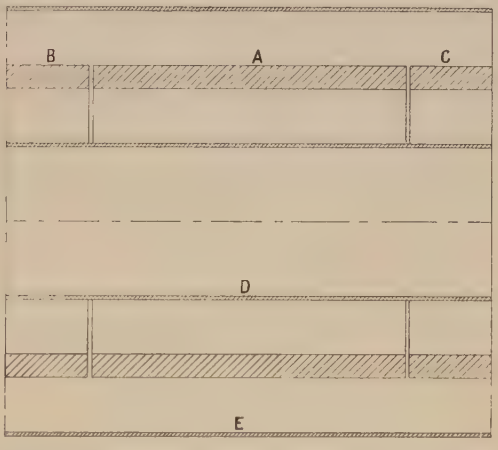

Fig. 4. The cross-sectional diagram of the electrode assembly. A, collecting electrode ; B, C, guard-rings ; D, E, inner and outer h.t. electrodes.

photographic plates interposed in the path of the forward neutrons. The track-lengths of recoiling protons were measured, and the energy spectrum obtained (fig. 3) was regarded as identical with the neutron energy spectrum. The maximum neutron energy agreed well with the calculated value.

The ionization chamber consisted essentially (fig. 4) of three annular cylindrical electrodes placed inside a vacuum-tight drum. The drum was filled with carbon tetrachloride vapour. The middle, collecting, electrode was fitted with guard-rings and was earthed through a resistance. The inner and outer

electrodes were maintained at +4 kv and +5·5 kv respectively. The sensitive volume consisted of two annular cylindrical spaces each 80 mm long and 14 mm in radial thickness. The inner and outer radii of the collecting electrode were 34 mm and 40 mm respectively. The maximum straight line path within the sensitive volume was 10·8 cm, which under the experimental conditions (saturated carbon tetrachloride vapour, 8·1 cm Hg pressure) corresponded to a residual energy of 1·5 MeV for protons and 5·5 MeV for α-particles. The effective fractional solid angle, that is that fraction of the emitted particles which could lose their entire energy within the sensitive volume, was a function of the path length, and was thus small for 1·5 MeV protons. It amounted to approximately 10^{-2} for α-particles of 4·25 MeV, the most energetic α-particles obtained in the experiment.

The chamber was filled by distillation under vacuum. While the chamber was open, liquid carbon tetrachloride was introduced into a reservoir attached to the drum and was then frozen. Air was pumped out and the carbon tetrachloride distilled into a glass U-tube. The pumping line was now sealed off and the carbon tetrachloride allowed to melt. The pressure of the carbon tetrachloride vapour was controlled by the temperature of the U-tube, while other parts of the chamber were maintained at a higher temperature; this also prevented the condensation of the vapour on the electrodes. The total pressure was measured on a mercury manometer, and the pressure of gases other than the carbon tetrachloride vapour, if any, was indicated by the difference of levels in the glass U-tube. This provided a convenient means of detecting failures of the vacuum system.

Difficulties were experienced in the design of the chamber on acconnt of the high voltage gradients required and the limited choice of materials. In gases exhibiting electron attachment reasonable collection times can only be obtained if relatively high fields are used. This, in turn, renders the use of insulators of high dielectric constant undesirable, as brushing occurs readily in the minute spaces between the electrodes and the insulating surfaces (Wilkinson 1950). In the present experiment the situation was complicated by the solvent properties of carbon tetrachloride which excluded the use of waxes and of most plastics. Further, sulphur could not be used as the Q-value of the reaction $^{32}S(n,\alpha)^{29}Si$ is positive. Eventually the electrodes were supported on Perspex. The supports were so located that protons knocked out of the material by the neutron flux could not enter the sensitive volume.

The pulses from the ionization chamber were fed into a preamplifier located next to the chamber. From there they passed on to a linear amplifier whose output was displayed on a cathode-ray oscillograph screen and recorded on film. The output was also connected to a monitoring cathode-ray oscillograph and to a scaler. The latter included a discriminator which enabled bias curves to be taken during the experiment. The high voltage for the h.t. electrodes of the ionization chamber was provided by a stabilized radio-frequency power supply. A pulse generator provided pulses of known and constant pulse height and rise time, for the purpose of monitoring the stability of gain of the system. The monitoring pulses were fed through a concentric condenser into the cable linking the ionization chamber and the preamplifier. The pulse generator was supplied from batteries and the stability was within 0·5%.

(iii) *Calibration*

A pulse-height–energy relation was found by placing a Th(B + C) source within the chamber, but outside the sensitive volume. The source could be moved in a line parallel to the axis of the chamber and was controlled from outside the vacuum system. The position of the source was adjusted until maximum pulse height was observed. This meant that the α-particles came to the end of their range at the end of the sensitive volume. If the α-particles stopped inside the sensitive volume the energy given up in it was obviously smaller; if they overshot the end of the sensitive volume by more than 1 mm air equivalent, the energy was also smaller since the specific ionization is greatest at the end of the range. The stopping power of the vapour was calculated from the range of the 8·78 MeV α-particles from ThC′ and from the knowledge of the position of the source. From the stopping power of the gas and from the length of the sensitive volume the energy given up in the sensitive volume was calculated as a function of the source position. Pulses obtained with two positions of the source were recorded on the film and were measured with the use of an optical projection arrangement.

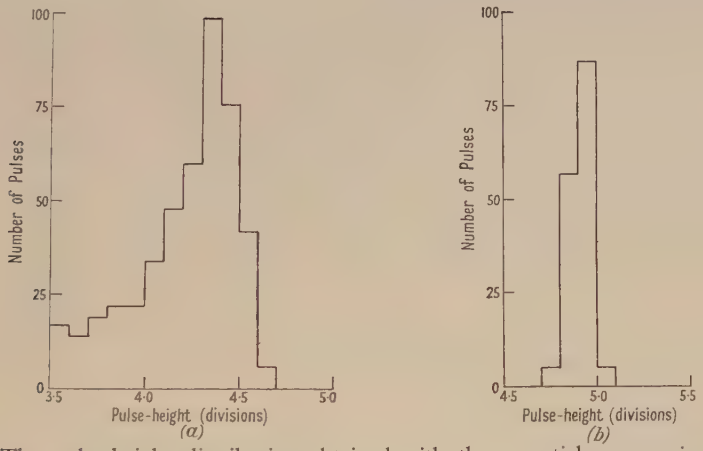

Fig. 5. (*a*) The pulse-height distribution obtained with the α-particle source in the position giving maximum pulse height; (*b*) The pulse-height distribution obtained with the pulses from the pulse generator. The spreading is due to noise in the amplifier.

The stopping power of the saturated carbon tetrachloride vapour at the pressure of 8·1 ± 0·05 cm Hg was found to be 0·38 ± 0·01 relative to air. This was in fair agreement with the value of 0·42 ± 0·04 calculated from the atomic stopping powers of carbon (Gray 1944) and chlorine (Geiger 1933). The energy given up by an α-particle crossing the sensitive volume and stopping at the end of it was calculated as 4·56 ± 0·09 MeV for a particle travelling in a direction parallel to the axis.

A large number of pulses were filmed with the Th(B + C) source in a position corresponding to this energy. Figure 5(*a*) shows the distribution obtained in terms of the divisions of the measuring scale. The general shape of the histogram corresponded to the energy spectrum of the source as measured before the source was introduced into the chamber. One division of the measuring scale was nearly equivalent to 1 MeV, so that for the purpose of the calculation of errors they could be regarded as exactly equivalent.

The effect of noise on the distribution of fig. 5 (*a*) was measured by feeding a number of constant height pulses from the pulse generator into the preamplifier. The resulting distribution is shown in fig. 5 (*b*). It will be seen that noise resulted in a probability of some 6% for a pulse to fall in the interval 0·1 Mev higher than that containing the most probable value. It followed that the real end-point of the calibration histogram (fig. 5 (*a*)) lay in the interval 4·6–4·7 divisions. Accordingly the scale calibration of 4·65 divisions for 4·56 ± 0·10 Mev was accepted.

Further, the effect of varying the differentiating and integrating time constants of the amplifier was investigated. Distributions of α-particle pulses were measured with the amplifier time constants set at 60, 100 and 140 microseconds (in each case the two time constants were equal). The position of the end-point of the distribution was found to be almost independent of the time constants in this range. This meant that the pulse heights were essentially independent of the location of the tracks of ionization in the chamber; it also showed that the controls of the amplifier could be safely re-set. The histogram of fig. 5 (*a*) was obtained with the setting of 60 μsec, used also in the rest of the experiment.

In the subsequent interpretation of results it was assumed that the pulse height varied proportionally with energy. This was justified by the end-point energy of the pulse height distribution obtained with the calibrating source withdrawn 30 mm away from the sensitive volume.

(iv) *Results*

Figure 6 (*a*) shows the total histogram obtained in the experiment. The pulse heights obtained in the individual experimental runs were corrected for small variations in the gain of the amplifier and the oscillograph. The pulses from the pulse generator were used for the purpose of this normalization.

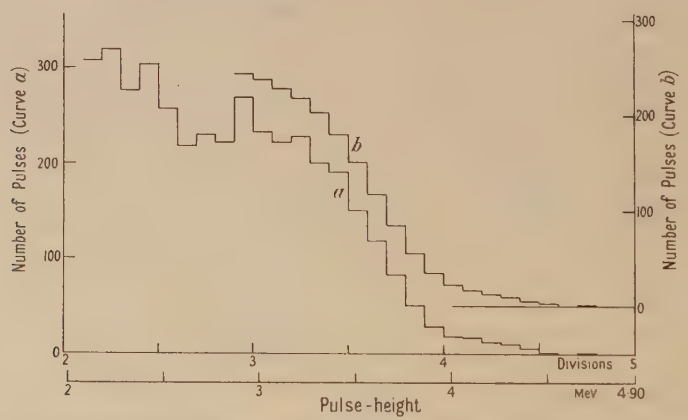

Fig. 6. (*a*) The experimental histogram of pulse heights due to α-particles from the reaction $^{35}Cl(n, \alpha)^{32}P$; (*b*) The same histogram after each ordinate had been broadened to show the effect of noise and 'piling up'.

The spread in the recorded pulse heights, obtained with the pulses from the pulse generator, was used to measure experimentally the joint effect of the amplifier noise and the 'piling up' of pulses. Figure 7 shows the distribution of pulse heights obtained when the output of the pulse generator was superimposed on the pulses arriving from the ionization chamber. The distribution includes only those pulses which were due to the pulse generator; these could be easily distinguished on the film, as they were regularly spaced in time.

In order to establish the effect of noise and 'piling up' on the location of the end-point of the histogram, the spreading represented in fig. 7 was applied to each ordinate of the histogram of fig. 6(*a*), thus producing the histogram of fig. 6(*b*). As can be seen this procedure made little difference to the appearance of the histogram, from which it was concluded that the histogram of fig. 6(*a*) represented truly the energy distribution of the pulses. It appeared possible, however, that the ordinate in the interval 4·5–4·6 divisions was due to spreading. Accordingly the end-point was taken as lying at $4{\cdot}5 \pm 0{\cdot}05$ divisions.

The expected shape of the histogram was computed theoretically on the basis of the geometry of the chamber, and on the assumptions that only one group of α-particles existed with a Q-value of $+1{\cdot}1$ MeV, that the reaction occurred isotropically, and that the incident neutron spectrum was uniform between the energies of 2·74 MeV and 3·34 MeV. The experimentally established spreading was applied to the computed histogram and the distribution of fig. 8 was thus obtained.

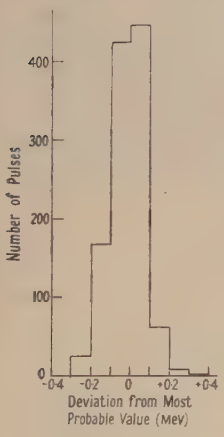

Fig. 7. The pulse-height distribution obtained with the pulses from the pulse generator superimposed on the pulses from the ionization chamber. The spreading is due to noise and to the 'piling up'.

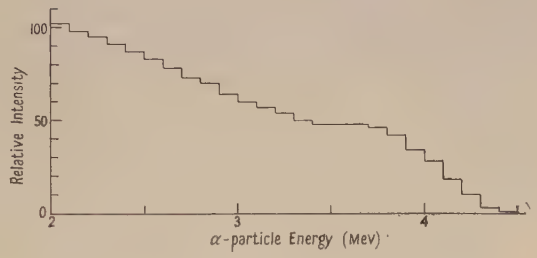

Fig. 8. The expected shape of the histogram.

The comparison of the shapes of the experimental (fig. 6(*a*)) and the expected (fig. 8) histograms provided an alternative way of establishing the position of the end-point and the probable error. This was done by means of a curve-fitting procedure, carried out for particles of energy greater than 3·8 MeV. The method confirmed the value of 4·5 divisions for the end-point, and gave a probable error of 0·1 division. The curve-fitting procedure consisted in choosing several possible positions for the end-point of the experimental histogram. In each case the relative weight of the histogram ordinates was computed as a function of the energy separation of the ordinate considered from the assumed end-point. The same procedure was carried out on the theoretical histogram, where the true value of the end-point energy was known. The theoretical weight distribution was compared with those obtained from the experimental histogram and that value of the end-point energy was accepted for which the best agreement was obtained.

The energy of the end-point of the histogram was therefore $4{\cdot}5 \pm 0{\cdot}1$ divisions or, converting to energy units and taking account of the probable error of the calibration, $4{\cdot}41 \pm 0{\cdot}15$ MeV. This pulse energy, together with the maximum neutron energy of 3·34 MeV, gave $Q = +1{\cdot}07 \pm 0{\cdot}15$ MeV for the Q-value of the reaction $^{35}\mathrm{Cl}(\mathrm{n}, \alpha)^{32}\mathrm{P}$.

It will be seen that the general shapes of the experimental and the computed histograms differ considerably. The experimental histogram indicates the presence of an unexpectedly large number of low energy α-particles. This may be interpreted as evidence of the presence of an α-particle group of lower energy. In view of the relatively small total number of particles, however, the evidence is considered inconclusive.

§4. CONCLUSION

The Q-values obtained for the reactions $^{32}S(\alpha, p)^{35}Cl$ and $^{35}Cl(n, \alpha)^{32}P$ are mutually consistent and the problem of the apparent discrepancy (see §1) in the mass difference of ^{32}S and ^{35}Cl is resolved. The value of the difference in mass excess derived from the reaction $^{32}S(\alpha, p)^{35}Cl$ is $1{\cdot}96 \pm 0{\cdot}11$ MeV. That derived from the reaction $^{35}Cl(n, \alpha)^{32}P$ and from the β-energy of ^{32}P is $1{\cdot}98 \pm 0{\cdot}15$ MeV. The two values agree well with the value obtained from Okuda's mass spectrographic measurements (see table).

Since the conclusion of the present author's experimental work a number of papers have appeared bearing on the question of the mass difference of ^{32}S and ^{35}Cl.

Collins, Nier and Johnson (1951) measured the masses of ^{32}S and ^{35}Cl, and obtained the values of $31{\cdot}982236 \pm (7 \times 10^{-6})$ M.U. and $34{\cdot}98004 \pm (5 \times 10^{-5})$ M.U. respectively. These figures gave $2{\cdot}04 \pm 0{\cdot}05$ MeV for the difference in mass excess of the two nuclei, which agrees within the experimental error with the result of the present author.

Wentink, Koski and Cohen (1951) determined spectroscopically the ratios of the mass differences of the sulphur isotopes ^{32}S, ^{33}S, ^{34}S and ^{35}S. From these ratios, with the use of Davison's (1949) values of the masses of ^{32}S, ^{33}S and ^{34}S, the authors calculated the mass of ^{35}S. The present author recalculated their results on the basis of the $^{32}S - ^{33}S$ mass difference, recently obtained by Strait *et al.* (1951) in a new investigation of the reaction $^{32}S(d, p)^{33}S$. The value thus obtained for the difference in mass excess of ^{32}S and ^{35}S was $2{\cdot}09 \pm 0{\cdot}33$ MeV. This taken in conjunction with the energy of the β-decay of ^{35}S (Cook *et al.* 1948) gave $2{\cdot}12 \pm 0{\cdot}33$ MeV for the $^{32}S - ^{35}Cl$ difference in mass excess, again in agreement with the present author.

The question remains open why the result of the present author disagrees with that of Metzger *et al.* (1947). It is perhaps significant to remark that the Q-value of the reaction $^{32}S(n, \alpha)^{29}Si$ was also measured in the Basel laboratory (Stebler and Huber 1948). The value obtained was $1{\cdot}16 \pm 0{\cdot}15$ MeV. The expected Q-value for this reaction can be calculated on the basis of the β-decay energy of ^{32}P and the Q-values of the reactions $^{31}P(d, \alpha)^{29}Si$ and $^{31}P(d, p)^{32}P$. The β-energy is $1{\cdot}712 \pm 0{\cdot}008$ MeV (Mattauch and Flammersfeld 1949), the first Q-value was measured as $+8{\cdot}158 \pm 0{\cdot}011$ MeV by Van Patter *et al.* (1952) and the second as $+5{\cdot}704 \pm 0{\cdot}009$ MeV by Strait *et al.* (1951). These figures lead to $Q = 1{\cdot}52 \pm 0{\cdot}02$ MeV for the reaction $^{32}S(n, \alpha)^{29}Si$. It thus appears that the values obtained in Basel for both the Q-value of the reaction $^{32}S(n, \alpha)^{29}Si$ and that of the reaction $^{35}Cl(n, \alpha)^{32}P$ are too low.

ACKNOWLEDGMENTS

The author wishes to thank Professor Sir George Thomson and Professor S. Devons for their help and for many valuable discussions.

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The M_{IV} and M_V Absorption Edges of ^{59}Pr, ^{60}Nd, ^{62}Sm and ^{63}Eu*

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Communicated by E. A. Stewardson; *MS. received 2nd May* 1952

ABSTRACT. Investigations on the M_{IV} and M_V absorption edges of four of the ' light ' rare earth elements are described, using a bent crystal focusing spectrometer. In particular, close attention has been paid to the occurrence of fine structure (' white lines ') on the long wavelength side of the edge. Such white lines are found at both edges for ^{59}Pr, ^{60}Nd, and ^{62}Sm, but for ^{63}Eu, while a white line is found at the M_V edge, it is absent or very weak at the M_{IV} edge.

§ 1. INTRODUCTION

IT has long been known that x-ray absorption edges of a number of elements are preceded on the long wavelength side by a narrow region of enhanced absorption giving the appearance on the photograph of a white line immediately before the main absorption edge. Thus Coster (1924) found such a line on the long wavelength side of the K edge of Mn in $KMnO_4$, while similar effects have been observed by Veldkampf (1935) for the L_{II} and L_{III} edges of Ta and W and by Sandström for the L_{II} and L_{III} edges of elements ranging from Sc to Cu. Among more recent examples of the effect, mention may be made particularly of the observation by Rule (1945) that a white line exists on the long wavelength side of the M_{IV} and M_V edges of Sm.

In the case of elements belonging to transition series it is natural to seek the origin of the 'white line' in transitions from the appropriate initial level into

* The material of this paper formed part of a thesis presented for the degree of M.Sc. at Birmingham University in December 1949.

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the incomplete electron groups characteristic of such elements. Such would seem to be the explanation of Rule's result for the rare earth element ^{62}Sm, where the vacant levels occur in the incomplete 4f shell relatively deep in the atom and probably well separated energetically from the continuum of levels outside the atom. As the final level in such white-line absorption would be in the 4f group, one would expect it to be well marked (owing to the operation of the usual selection rules) in the case of the M_{IV} and M_V edges, and such effects should be in evidence more or less for the whole series of the rare earth elements. The present paper describes the results of investigations into the M_{IV} and M_V edges of four of the 'light' rare earth elements, namely ^{59}Pr, ^{60}Nd, ^{62}Sm and ^{63}Eu. Of the four elements mentioned only one, ^{62}Sm, had previously been investigated, as stated above, by Rule. All four elements have been used in the form of oxide only.

§2. APPARATUS AND EXPERIMENTAL METHOD

The M_{IV} and M_V absorption edges of the rare earths lie in the soft x-ray region (from *c.* 8 Å upwards), and a vacuum spectrograph has been specially constructed for the investigation. The spectra are produced by a bent crystal focusing spectrometer employing the reflection method of Johann (1931). The general design owes much to the various published descriptions of focusing x-ray spectrometers, particularly those of Cauchois (1932, 1933, 1945) and to information supplied privately by her.

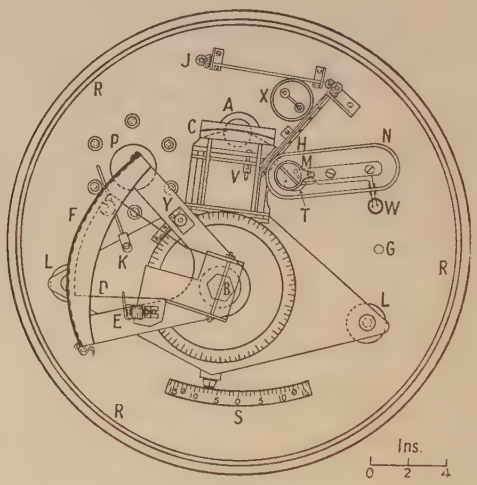

Fig. 1. A, pivot for spectrometer ; B, pivot for film holder ; C, crystal holder ; D, selector shutter ; E, electromagnet for operating shutter ; F, film holder ; G, to McLeod gauge ; H, Al foil carrier ; J, ground joint ; K, spectrometer clamp ; L, levelling screw ; M, filament assembly ; N, lead screen ; P, pumping port ; R, groove for rubber gasket ; S, scale ; T, target ; V, adjusting screw for crystal holder ; W, insulated leads for filament ; X, evaporation oven ; Y, film-holder clamp.

The whole system, consisting of x-ray source, evaporation plant for preparation of absorption films and spectrometer, is housed in a mild-steel cylindrical tank. A plan showing the disposition of the various components on the base plate of the tank is shown in fig. 1. The crystal used to produce the spectra is mica bent to a radius of curvature of 16 in., giving a focal circle of 8 in. radius. This

small radius necessitates the use of photographic film instead of plate. The dispersion is approximately 40 X.U. per mm at 10000 X.U. The X-ray source is of conventional hot-cathode and target type and gives a line source about 15 mm long situated about 8 cm from the crystal. The water-cooled anticathode has three targets—one of tungsten for production of continuous spectra, and two of copper which carry materials for producing reference spectra. The cathode is made of oxide-coated nickel strip to avoid production of excessive light within the tank. This strip is replaced after each exposure.

Absorption films of the rare earth oxide are made by evaporating the material on to thin aluminium foil from an electrically heated molybdenum or tantalum strip. The oven is fastened to the under side of the base plate and, after receiving its coating, the aluminium foil can be brought into the X-ray beam between the source and crystal by means of a mechanical device operated from outside the tank. The absorption film is thus prepared and brought into position without being exposed to the atmosphere. The tank is exhausted by a mercury diffusion pump which maintains a pressure of the order of 10^{-2} micron.

The exposures needed to register the absorption spectra range from 300 to 1240 milliamp-hours. With currents of the order of 25 mA this means actual exposure times up to 50 hours. Each exposure is made continuously without any intermission. High-order K spectra of elements ranging from ^{15}P to ^{26}Fe are used as reference spectra.

Wavelengths are determined by the now standard procedure of interpolating linearly between two known reference lines to determine the Bragg angle of the required line. The wavelength of this line or point in the spectrum is then derived in the usual fashion from the Bragg angle so computed. The wavelengths of reference lines are taken from the tables of Cauchois and Hulubei (1947) and results are therefore expressed in the units used in those tables, i.e. X.U. The computation of wavelengths involves a knowledge of the appropriate lattice constant of the crystal used, in this case mica. The lattice constant of mica however is known to be slightly variable. In some preliminary experiments the wavelengths of a number of known lines, particularly L lines of tungsten, were determined using the values for mica given by Siegbahn (1931), multiplying his values in ångströms by 1000 and treating the result as X.U. This was found to give the values of the wavelengths of the known lines differing from the values tabulated in the tables of Cauchois and Hulubei by amounts less than the probable experimental error. These values for the lattice constants of mica have therefore been used throughout. This procedure is justified by the excellent agreement obtained between Rule's measurements on ^{62}Sm and the values found in this work.

For the determination of the wavelengths of lines, readings of positions taken directly from the negative are probably adequate, but for the determination of the position of absorption edges it is advisable to make measurements on the density curves in their neighbourhood. As a microphotometer was not available in the laboratory at the time, earlier measurements were made with a non-recording microphotometer in the laboratories of the British Thomson-Houston Company at Rugby, while final measurements have been made from curves obtained with the recording microphotometer in the Physical Laboratories of the Imperial College of Science, London. The overall error in wavelength determination is estimated to be about ± 2 X.U.

The thickness of the absorption films is estimated from the amount of rare earth oxide evaporated and the distance from the source to the foil on which deposition took place. This gives results reliable to order of magnitude only, but on the other hand the relative values of the thicknesses are probably fairly trustworthy as the evaporations are carried out under as nearly as possible identical conditions. The greatest uncertainty exists in the case of ^{59}Pr as this is used in the form of the higher oxide Pr_6O_{11}, and it is possible that it loses some part of its oxygen in the process of evaporation, reverting to some extent to Pr_2O_3.

The question of the optimum thickness of absorption film needed to give best contrast at the absorption edge has been considered by Sandström (1930) using Jönssen's absorption law (1928), while McGrath (1939) has worked out the optimum thickness in the case of gold. For the M_{IV} and M_V edges he gives 0·50 and 0·79 mg/cm^2 respectively. Thicknesses of the order of one-fifth of these values are found most suitable for bringing out the white lines accompanying the M_{IV} and M_V edges of the rare earths described in the present paper.

§ 3. RESULTS

'White line' structure is found on the long wavelength side of nearly all the edges studied, the only case where it appears to be absent, or at least not prominent, being the M_{IV} edge of Eu. This type of structure is very clear to the naked eye on the negatives themselves. Photometer curves of the negatives have been taken in all cases, rather with a view to determining more accurately the positions of maxima and minima than to reveal structure. In actual fact the photometer curves reveal no structure that is not readily noticeable to the eye.

In many cases 'Lindsay–Kronig' structure is noticeable on the short wavelength side of the edges. This is, in general, much fainter than the white line on the long wavelength side of the edge. No attempt has been made to determine the optimum conditions for bringing it out, and it is ignored in the present account of the investigation.

Tracings of the photometer curves obtained for the various edges investigated are shown in figs. 2–8, while wavelengths of the various points of possible physical significance on the curves are given in tables 1–4 (all wavelengths are given in x.u.). In referring to the curves the following symbols are used: K (with subscript if necessary) is used to indicate a point of inflection; other roman capitals denote minima (i.e. absorption *maxima*) and Greek letters the intervening maxima (i.e. absorption minima).

The figures show the results obtained for absorption films of four different thicknesses each of Sm and Nd, one of Pr and two of Eu. In order to link up with the only previous work in this field (that of Rule 1945) a study of the samarium absorption was undertaken first. It is satisfying to find close agreement with Rule's results in this case, although he distinguishes additional components (two on each side of B) which have not been observed in the present investigation (due possibly to the use of a rather smaller dispersion). In giving his results for comparison therefore the mean of his two components is given in brackets and equated to the wavelength of the single point of inflection found in the present work. In this and all other tables gaps in the list of wavelengths signify that the corresponding points on the curves are not sufficiently well defined for accurate wavelength determination.

In all cases the following general features are noticeable: for very thin absorption films the absorption over the whole region of the edge appears almost line-like in character, but changes gradually to the typical edge-like absorption as the thickness is increased. The white lines, when present (indicated by the minima B, C . . .), appear to move down the edge as thickness increases, becoming merged eventually in the general absorption of the edge, so that for very great thickness the 'edge' appears normal, but is actually represented by the long wavelength side of the line. The wavelengths of the lines and points of inflection, however, show no systematic change with thickness—a satisfactory result, indicating

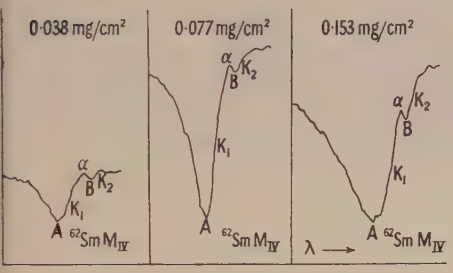

Fig. 2.

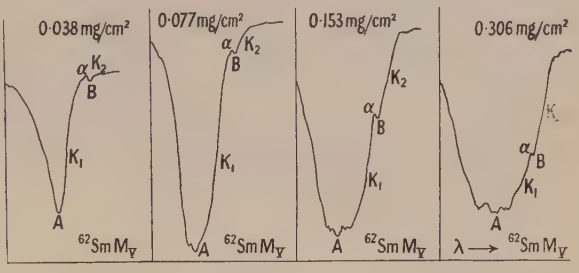

Fig. 3.

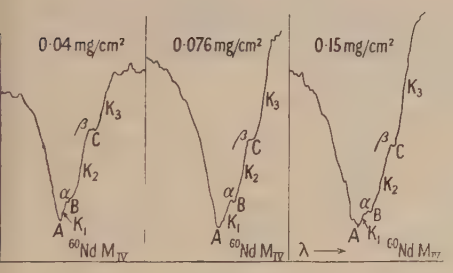

Fig. 4.

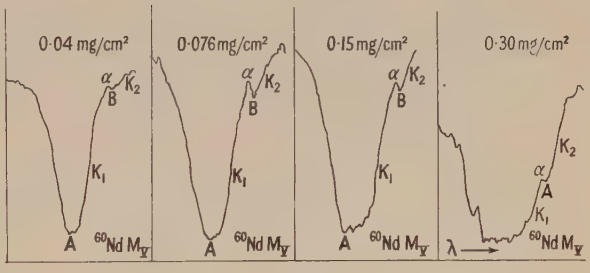

Fig. 5.

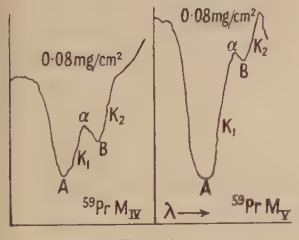

Fig. 6.

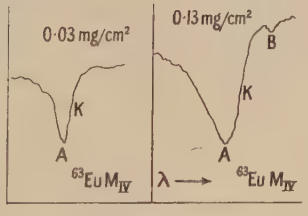

Fig. 7.

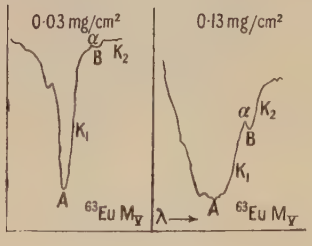

Fig. 8.

that the films are throughout still sufficiently thin to make negligible any difference between turning points on the curve of blackening and those of the true absorption curve (cf. Sandström 1936). In order to save space, in tables 1–4 the mean values only for the wavelengths are given, these being averaged over the various thicknesses of absorbing film.

Turning next to particular results, it will be seen that there is positive evidence for the occurrence of (*a*) one white line at both M_{IV} and M_V edges of ^{59}Pr, (*b*) two white lines at the M_{IV} edge of ^{60}Nd and one at the M_V edge, (*c*) one at

both edges of ^{62}Sm, and (*d*) one at the M_V edge of ^{63}Eu. With regard to the last-mentioned element there is no sign of a white line at the M_{IV} edge for the thinner film (0·03 mg/cm²), but for the thicker film (0·13 mg/cm²) there is a doubtful

Table 1. M_{IV} and M_V edges of ^{62}Sm

	M_{IV} edge			M_V edge		
	λ (mean)	λ (Rule)	$\frac{\nu}{R}$ (mean)	λ (mean)	λ (Rule)	$\frac{\nu}{R}$ (mean)
K_2	11262·6	(11260·5)	80·91	11525·6	(11525·2)	79·06
B	11250·5	11250·4	80·998	11510·6	11511·1	79·16
α	11242·5	11240·1	81·06	11505·2	11500·5	79·21
K_1	11224·2	(11228·0)	81·187	11489·2	(11487·9)	79·315
A	11200·7	11203·2	81·36	11454·2	11460·1	79·56

Table 2. M_{IV} and M_V edges of ^{60}Nd

	M_{IV} edge		M_V edge	
	λ (mean)	$\frac{\nu}{R}$ (mean)	λ (mean)	$\frac{\nu}{R}$ (mean)
K_3	12433	73·29		
C	12415	73·401		
β	12411	73·42		
K_2	12398	73·58	12711	71·69
B	12390	73·549	12696	71·776
α	12386	73·572	12689	71·82
K_1	12377	73·626	12666	71·946
A	12364	73·703	12633	72·13

Table 3. M_{IV} and M_V Edges of ^{59}Pr

	M_{IV} edge		M_V edge	
	λ (mean)	$\frac{\nu}{R}$ (mean)	λ (mean)	$\frac{\nu}{R}$ (mean)
K_2	13095	69·589	13366	68·178
B	13075	69·695	13354	68·239
α	13061	69·770	13349	68·265
K_1	13042	69·872	13330	68·362
A	13019	69·993	13310	68·465

Table 4. M_{IV} and M_V Edges of ^{63} Eu

	M_{IV} edge			M_V edge	
	λ (mean)	$\frac{\nu}{R}$ (mean)		λ (mean)	$\frac{\nu}{R}$ (mean)
K	10689	85·25	K_2	10990	82·92
A	10667	85·43	B	10981	82·99
			α	10970	83·07
			K_1	10949	83·23
			A	10931	83·37

trace of a white line at this edge (shown at B in fig. 7). It is mentioned with some reserve as it is very faint and has been observed in this one instance only. Its wavelength is approximately 10 730 X.U. The existence of the line however is certainly in need of confirmation.

§ 4. DISCUSSION

The common occurrence of the white lines for all four elements studied supports the general idea that they arise from transitions from the $M_{IV, V}$ groups to the incomplete 4f ($N_{VI, VII}$) groups, and, taking account of the fact that the selection rules would 'forbid' only the $M_{IV} \rightarrow N_{VII}$ transition, the occurrence of prominent white lines at both edges for ^{59}Pr, ^{60}Nd and ^{62}Sm but at the M_V edge only for ^{63}Eu suggests that the lighter rare earths (^{58}Ce to ^{62}Sm) tend to build up an N_{VI} electron group, this group becoming complete at ^{63}Eu, in general conformity with the simplest possible picture of the building up of the rare earth series (see e.g. v. Hevesy 1927). It would seem unwise to stress this, however, until data about other members of the series are available.

Considering next the true edges, these should arise from transitions from $M_{IV, V}$ to the continuum of unoccupied levels commencing at a higher value than the $N_{VI, VII}$ levels—possibly $O_{VI, VII}$. Granting the above interpretation of the white lines, it follows that the edges should be represented approximately by the points of inflection between the point of maximum absorption (A in all cases) and the first white line which occurs on the long wavelength side of this maximum. That is to say, the M_{IV} edges correspond to the points K_1 for ^{59}Pr, ^{60}Nd and ^{62}Sm and to K for ^{63}Eu, and the M_V edges to the K_1 for all four elements. These points of inflection will not give the true wavelength of the edge, however, as the absorption of the neighbouring white line will be partly superimposed on it and a correction should be applied (cf. Sandström 1935). This correction has not been made in the present instance as in general the profiles of the white lines have not been well enough brought out. A rough estimate shows that it may increase the actual wavelengths by about 5 X.U.

In his paper on the samarium edges Rule seeks to establish correspondence between the physically significant points on the aborption curves with the M-emission lines as measured by Lindberg (1931). As there is quite close agreement between Rule's wavelengths and those found here, his correspondences may be regarded as to some extent confirmed, although in general it is not clear what physical significance should be attached to such 'correspondence'. The fairly close agreement between the white line minimum A on the M_{IV} edge (11 250·4 X.U.) and the $M\beta_{II}$ emission line (11 254 X.U., ascribed to the transition M_{IV}–N_{VI}) may, however, be of significance. In the case of Nd and Eu a comparison of Lindberg's emission results with the results given in this paper have revealed no obvious correspondences. For Pr no emission data for the M series seem to have been published. An attempt has therefore been made to photograph the M spectrum of this element. This has proved to be fairly difficult. While the $M\beta$ line can be obtained quite readily, it is very difficult to obtain the α line with any intensity. Trouble is also experienced with the rather intense L radiation of the copper target, which tends to overlap and obscure the Pr lines. Some exposures have therefore been made using a molybdenum target as a support for the Pr oxide. This allows the lines to be brought out sufficiently well to be measured, with some uncertainty, however, as they are very diffuse. Using Cl–Kα_1 (3rd order) and Mo–L$\gamma_{2,3}$ (3rd order) as reference lines the following values for the wavelengths are obtained: Mα, 13 315 X.U.; Mβ_I, 12 977 X.U.; Mβ_{II}, 13 022 X.U.; Mβ_{III}, 13 063 X.U. Here again no obvious or close correspondence seems to exist between either of the white lines observed for Pr and any of the emission lines listed above.

ACKNOWLEDGMENTS

This work was carried out in the Physics Department of the University College of Leicester under the supervision of Professor E. A. Stewardson. I am indebted to him for suggesting the problem and for his advice and encouragement throughout. My thanks are due also to the British Council for the award of a scholarship during part of the period in which the work was carried out. I would express my grateful thanks to Professor Y. Cauchois for much helpful advice, particularly on X-ray spectrometers, and for allowing me to see spectrometers designed and used by her in her laboratory in Paris. I am indebted also to the British Thomson-Houston Company, Rugby, for the use of their non-recording microphotometer and to Sir George Thomson and Professor R. W. B. Pearse for the use of the recording microphotometer at Imperial College, London. Finally, I should like to express my gratitude to Mr. N. A. Hunt for valuable suggestions in the practical design of the spectrometer and for his skill in carrying out its construction in the workshop of the Physics Department of the University College of Leicester.

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The Second Virial Coefficient near Absolute Zero

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MS. received 3rd June 1952

ABSTRACT. By the solution of Bloch's equation for the density matrix, a development is obtained suitable for the exact evaluation of the second virial coefficient near absolute zero. Assuming a molecular interaction of the Lennard-Jones type, it is shown that the virial coefficient for helium is $-\{\frac{1}{2}(\pi\beta)^{3/2}+31{\cdot}15\,\pi\beta+\mathrm{O}(1)\}\text{Å}^3$, $\pi\beta=38{\cdot}04/T$.

General formulae are given for fluids the molecules of which satisfy Bose or Fermi statistics.

§1. INTRODUCTION

THE second virial coefficient, which measures the deviation of slightly dense gases from the classical perfect gas law, is a physical quantity of considerable importance. As shown by Lennard-Jones (1924), a comparison of theoretical and experimental values may be used to infer the

intermolecular force law. In the case of helium it was possible to check the forces so obtained by Buckingham (1938) with those derived by Slater and Kirkwood (1931) from the known structure of the helium atom. A small but noticeable discrepancy could be attributed to quantum-mechanical effects, neglected in Lennard-Jones' treatment, and was largely removed by applying a quantum-mechanical correction, in the manner of de Boer and Michels (1938).

The quantum-mechanical correction to the second virial coefficient was evaluated by Uhlenbeck and Beth (1936, 1937) by a method which is effective at temperatures above 20° K. Taking account of quantum statistics, Massey and Buckingham (1938), Gropper (1939), de Boer (1940), and Kahn (1938) later attempted to extend calculations to lower temperatures, but their methods also fail in the interesting region below 2° K. The purpose of the present paper is to determine values of the second virial coefficient in the neighbourhood of 0° K, for fluids whose molecules satisfy Bose or Fermi statistics. This is, so far as the author is aware, the first rigorous theoretical calculation of any physical quantity at liquid helium temperatures, based on a realistic molecular model. The result may be useful in low temperature thermometry, which has so far had to rely on the indirect method of adiabatic demagnetization at temperatures below 1° K.

The method is a specialization of one developed recently by the author (Green 1952) which leads to formulae from which all the virial coefficients might, in principle, be calculated. However, since the example considered is much simpler than the general case, the theory required is here developed briefly from first principles. The point of departure is Bloch's equation for the density matrix; one could, however, use other methods—for example, the operational method of Goldberger and Adams (1952), or that suggested by the author (Green 1951) starting from an equation due to Wigner (1932). The common feature of these methods is a development in terms of the force constant. The convergence of this development is here established in the most convincing way, by the summation of the resulting series.

The intermolecular force law adopted is that which gives a potential energy

$$\begin{aligned} \phi(r) &= -\lambda r^{-6} + \mu r^{-10}, & r \geqslant \epsilon \\ &= \phi(\epsilon), & r < \epsilon \end{aligned} \qquad \ldots\ldots(1)$$

between the two molecules. For reasons well known in quantum theory, difficulties arise in dealing with very singular potential functions, which require the introduction of the small quantity ϵ; however, on physical grounds one would not expect results to depend very sensitively on this parameter, and in fact it is possible to determine an asymptotic value of the second virial coefficient in the limit $\epsilon = 0$.

An interesting difference emerges between the virial coefficients of fluids which satisfy Bose statistics, and those which satisfy Fermi statistics. In the former case, the deviation of the second virial coefficient from that of an ideal gas is inversely proportional to the temperature near absolute zero; in the latter case it approaches a constant value. In each case, the terms arising from the interaction of the molecules play a comparatively important part above 0·1° K.

Little support is found for the contention (Matsubara 1951) that quantum statistics play a more essential role than quantum mechanics in determining the behaviour of fluids at very low temperatures. Classical statistics occupy a position intermediate between the two kinds of quantum statistics; but the application of quantum mechanics is always essential.

§2. SOLUTION OF BLOCH'S EQUATION

For a system of two equal particles whose mutual potential energy at relative displacement $\mathbf{r}$ is $\phi(\mathbf{r})$, Bloch's equation for the density matrix is

$$\left(\frac{\hbar^2}{m}\frac{\partial^2}{\partial \mathbf{r}^2}+\frac{\hbar^2}{4m}\frac{\partial^2}{\partial \mathbf{x}^2}\right)\rho=\left\{\frac{\partial}{\partial\beta}+\phi(\mathbf{r})\right\}\rho \qquad \ldots\ldots(2)$$

where $\beta=1/(kT)$ and $\mathbf{x}$ is the coordinate vector of the mass-centre. Units of mass and time will henceforth be chosen so that $\hbar=m=1$.

The density matrix ρ is subject to the boundary condition

$$\rho(\mathbf{x},\mathbf{r};\mathbf{x}',\mathbf{r}')=\delta(\mathbf{r}-\mathbf{r}')\delta(\mathbf{x}-\mathbf{x}') \qquad \text{for} \qquad \beta=0 \qquad \ldots\ldots(3)$$

in classical statistics; in quantum statistics one has

$$\rho_{\pm}(\mathbf{x},\mathbf{r};\mathbf{x}',\mathbf{r}')=\rho(\mathbf{x},\mathbf{r};\mathbf{x}',\mathbf{r}')\pm\rho(\mathbf{x},-\mathbf{r};\mathbf{x}',\mathbf{r}'), \qquad \ldots\ldots(4)$$

where the plus sign distinguishes Bose statistics, and the minus sign Fermi statistics.

The solution of (2) which satisfies the initial condition (3) is

$$\rho=(\pi\beta)^{-3/2}\exp\{-(\mathbf{x}-\mathbf{x}')^2/\beta\}\,\sigma(\mathbf{r},\mathbf{r}') \qquad \ldots\ldots(5)$$

where

$$\sigma(\mathbf{r},\mathbf{r}')=\mathrm{e}(\mathbf{r}-\mathbf{r}',\beta)-\int_0^{\beta}d\beta_1\int d\mathbf{r}_1\phi(r_1)\mathrm{e}(\mathbf{r}-\mathbf{r}_1,\beta-\beta_1)\left\{\mathrm{e}(\mathbf{r}_1-\mathbf{r}',\beta_1)\right.$$
$$\left.-\int_0^{\beta_1}d\beta_2\int d\mathbf{r}_2\phi(r_2)\mathrm{e}(\mathbf{r}-\mathbf{r}_1,\beta-\beta_1)\mathrm{e}(\mathbf{r}_1-\mathbf{r}_2,\beta_1-\beta_2)\mathrm{e}(\mathbf{r}_2-\mathbf{r}',\beta_2)\right\}-\ldots, \qquad \ldots\ldots(6)$$

$$\mathrm{e}(\mathbf{r},\beta)=(4\pi\beta)^{-3/2}\exp(-\mathbf{r}^2/4\beta).$$

This is readily verified by direct substitution, once it has been observed that $\mathrm{e}(\mathbf{r}-\mathbf{r}',\beta-\beta')$ is the solution of (2) with $\phi(r)=0$, which approaches $\delta(\mathbf{r}-\mathbf{r}')$ as β' approaches β from below. The integrations with respect to β_1, β_2, etc. in (6) could be carried out immediately with the help of the mathematical identity*

$$\int_0^{\beta}d\beta_1\mathrm{e}(\mathbf{s},\beta-\beta_1)\mathrm{e}(\mathbf{t},\beta_1)=\frac{(s+t)}{4\pi st}\,\frac{\exp\{-(s+t)^2/4\beta\}}{(4\pi\beta)^{3/2}}. \qquad \ldots\ldots(7)$$

However, these integrations are better left till later.

§3. THE SECOND VIRIAL COEFFICIENT

The second virial coefficient is given by the formula

$$B=\tfrac{1}{2}\int\{1-g(r)\}\,d\mathbf{r} \qquad \ldots\ldots(8)$$

where $g(r)$ is the probability of finding two molecules, unperturbed by any others, at distance r, normalized to 1 at large distances. The quantum-mechanical derivation of this formula is contained in the quantum-theoretical generalization of Mayer's cluster theory by Uhlenbeck and Kahn (1938). Since

$$g(r)=(2\pi\beta)^3\rho(\mathbf{x},\mathbf{r};\mathbf{x},\mathbf{r}), \qquad \ldots\ldots(9)$$

* Reduced by substitution $\beta_1=\beta u^2/(1+u^2)$ to the integral (021·10) of Groebner and Hofreiter's *Integraltafel*, vol. 2.

the second virial coefficient for quantum statistics is given by

$$\left.\begin{aligned} B_{\pm} &= B \pm B' \\ B &= \tfrac{1}{2}\int \{1-(2\pi\beta)^3\rho(\mathbf{x},\mathbf{r};\mathbf{x},\mathbf{r})\}\,d\mathbf{r}, \\ B' &= -\tfrac{1}{2}(2\pi\beta)^3\int \rho(\mathbf{x},-\mathbf{r};\mathbf{x},\mathbf{r})\,d\mathbf{r}, \end{aligned}\right\} \quad \ldots\ldots(10)$$

if B always represents the value for classical statistics.

Making use of (5) and (6), and also the identity

$$\int d\mathbf{r}\mathrm{e}(\mathbf{r}-\mathbf{r}_1,\beta-\beta_1)\mathrm{e}(\mathbf{r}_2-\mathbf{r},\beta_2)=\mathrm{e}(\mathbf{r}_2-\mathbf{r}_1,\beta-\beta_1+\beta_2) \quad \ldots\ldots(11)$$

one obtains

$$(4\pi\beta)^{-3/2}\left\{2B/\beta-\int d\mathbf{r}_1\phi(\mathbf{r}_1)\right\}=-\tfrac{1}{2}\int_0^\beta d\beta_1\int d\mathbf{r}_1\phi(\mathbf{r}_1)\int d\mathbf{r}_2\phi(\mathbf{r}_2)$$
$$\times\mathrm{e}(\mathbf{r}_2-\mathbf{r}_1,\beta-\beta_1)\mathrm{e}(\mathbf{r}_1-\mathbf{r}_2,\beta_1)+\tfrac{1}{3}\int_0^\beta d\beta_1\int_0^{\beta_1}d\beta_2\ldots. \quad \ldots\ldots(12)$$

The integrations with respect to β_1, β_2, etc. are now performed with the help of (7), giving

$$2B/\beta=\int\phi(\mathbf{r}_1)\,d\mathbf{r}_1-\int d\mathbf{r}_1\phi(\mathbf{r}_1)\int\frac{d\mathbf{r}_2\phi(\mathbf{r}_2)}{4\pi|\mathbf{r}_2-\mathbf{r}_1|}\Big[\exp\{-4(\mathbf{r}_2-\mathbf{r}_1)^2/4\beta\}$$
$$-\int\frac{d\mathbf{r}_3\phi(\mathbf{r}_3)}{4\pi|\mathbf{r}_3-\mathbf{r}_2|}\exp\{-(|\mathbf{r}_2-\mathbf{r}_1|+|\mathbf{r}_3-\mathbf{r}_2|+|\mathbf{r}_1-\mathbf{r}_3|)^2/4\beta\}\Big]-\ldots. \quad \ldots\ldots(13)$$

Similarly

$$2B'\beta=-(\pi\beta)^{3/2}/\beta+\int d\mathbf{r}_1\phi(r_1)\Big[\exp(-4\mathbf{r}_1{}^2/4\beta)$$
$$-\int\frac{d\mathbf{r}_2\phi(r_2)}{4\pi|\mathbf{r}_2-\mathbf{r}_1|}\exp\{-(|\mathbf{r}_2-\mathbf{r}_1|+|\mathbf{r}_1+\mathbf{r}_2|)^2/4\beta\}\Big]+\ldots. \quad \ldots\ldots(14)$$

For large values of β the right-hand sides of these expressions can be developed in series of the type

$$\left.\begin{aligned} 2B/\beta &= R_0+R_1\beta^{-1}+R_2\beta^{-2}+\ldots., \\ 2B'/\beta &= R_0'+R_1'\beta^{-1}+\ldots.-(\pi\beta)^{3/2}/\beta, \end{aligned}\right\} \quad \ldots\ldots(15)$$

where $R_0=R_0'=\int\phi(r)\tau(r)\,d\mathbf{r}$,

$$\tau(r)=1-\int\frac{d\mathbf{r}_1\phi(r_1)}{4\pi|\mathbf{r}-\mathbf{r}_1|}+\int\frac{d\mathbf{r}_1\phi(r_1)}{4\pi|\mathbf{r}-\mathbf{r}_1|}\int\frac{d\mathbf{r}_2\phi(r_2)}{4\pi|\mathbf{r}_2-\mathbf{r}_1|}-\ldots. \quad \ldots\ldots(16)$$

and

$$R_1=\tfrac{1}{4}\int d\mathbf{r}\phi(r)\int d\mathbf{r}'\phi(r')\{3|\mathbf{r}-\mathbf{r}'|\tau(r)\tau(r')/4\pi+|\mathbf{r}-\mathbf{r}'|^2\kappa(\mathbf{r},\mathbf{r}')\}$$
$$-2\pi\left\{\int\phi(r)\tau(r)\,d\mathbf{r}/4\pi\right\}^3,$$

$$\kappa(\mathbf{r},\mathbf{r}')=\frac{1}{4\pi|\mathbf{r}-\mathbf{r}'|}-\int\frac{d\mathbf{r}_1\phi(r_1)}{4\pi|\mathbf{r}-\mathbf{r}_1|4\pi|\mathbf{r}_1-\mathbf{r}'|}+\ldots. \quad \ldots\ldots(17)$$

while $$R_1-R_1'=\int\phi(r)\mathbf{r}^2\,d\mathbf{r}-\int d\mathbf{r}\phi(r)\int d\mathbf{r}'\phi(r')\mathbf{r}\,.\,\mathbf{r}'\kappa(\mathbf{r},\mathbf{r}'). \quad \ldots\ldots(18)$$

The higher coefficients are of the type

$$R=R_1+R_2\ln\beta+R_3\beta^{1\,2}+R_4\beta^{1/2}\ln\beta \quad \ldots\ldots(19)$$

as one sees readily by making a development of a typical integral

$$\int_0^\infty \phi(r)\{\exp(-r^2/\beta)\}r^n\,dr$$

for large values of β, and integral values of n. It is clear, however, that an evaluation of R_0, R_1, R_0' and R_1' will be sufficient to determine the second virial coefficient, near absolute zero.

It is not immediately obvious that the infinite series which define $\tau(r)$ and $\kappa(\mathbf{r}, \mathbf{r}')$ in (16) and (17) converge. Assuming this for the present, they then satisfy the integral equations

$$\tau(r) = 1 - \int \frac{\tau(r_1)\phi(r_1)\,d\mathbf{r}_1}{4\pi|\mathbf{r}-\mathbf{r}_1|} \qquad \ldots\ldots(20)$$

and

$$\kappa(\mathbf{r}, \mathbf{r}') = \frac{1}{4\pi|\mathbf{r}-\mathbf{r}'|} - \int \frac{\kappa(\mathbf{r}_1, \mathbf{r}')\phi(r_1)\,d\mathbf{r}_1}{4\pi|\mathbf{r}-\mathbf{r}_1|}. \qquad \ldots\ldots(21)$$

In the next section, it will be shown that these integral equations possess unique solutions, which can be expanded as double power series in λ and μ. These solutions must be the sums of the series defining $\tau(r)$ and $\kappa(\mathbf{r}, \mathbf{r}')$, which therefore converge.

As a matter of interest, it may be noted that

$$\tau(r) = 1 - \int \kappa(\mathbf{r}, \mathbf{r}')\phi(r')\,d\mathbf{r}'. \qquad \ldots\ldots(22)$$

§4. SOLUTION OF THE INTEGRAL EQUATIONS

The integral equation (20) is solved by first noticing that $\tau(r)$ satisfies the differential equation

$$\frac{1}{r}\frac{d^2}{dr^2}\{r\tau(r)\} = \frac{\partial^2}{\partial \mathbf{r}^2}\tau(r) = \phi(r)\tau(r) \qquad \ldots\ldots(23)$$

and also the boundary conditions

$$\tau(r) = 1 \quad \text{when} \quad r = \infty, \qquad r\tau(r) = 0 \quad \text{when} \quad r = 0. \qquad \ldots\ldots(24)$$

Substituting (1) into (22), one has

$$r\tau(r) = A_0 \sinh[\{\phi(\epsilon)\}^{1/2}r] \qquad r \leqslant \epsilon \qquad \ldots\ldots(25)$$

where A_0 is a constant; also, since the equation reduces to

$$\frac{d^2}{dz^2}\{z^{3/8}\tau\} + (-\tfrac{1}{4} + kz^{-1} + \tfrac{15}{64}z^{-2})z^{3/8}\tau, \qquad \ldots\ldots(26)$$

$$z = \tfrac{1}{2}\mu^{1/2}r^{-4}, \qquad k = \tfrac{1}{8}\lambda\mu^{-}\epsilon^{1/2}$$

for $r \geqslant \epsilon$, the solution in terms of the confluent hypergeometric function must be*

$$\tau = z^{-3/8}\{M_{k,-\frac{1}{8}}(z) + A(\tfrac{1}{2}\mu^{1/2})^{-1/4}M_{k,\frac{1}{8}}(z)\}, \qquad r \geqslant \epsilon. \qquad \ldots\ldots(27)$$

The constants A_0 and A in (25) and (27) must be chosen to ensure the continuity of $\tau(r)$ and $\tau'(r)$. Thus the value of A is determined by

$$\epsilon\{\phi(\epsilon)\}^{1/2}\coth[\epsilon\{\phi(\epsilon)\}^{1/2}] = -\tfrac{1}{4}z_0 f'(z_0)/f(z_0), \qquad z_0 = \tfrac{1}{2}\mu^{1/2}\epsilon^{-4},$$

$$f(z) = z^{-5/8}\{(\tfrac{1}{2}\mu^{1/2})^{1/4}M_{k,-\frac{1}{8}}(z) + AM_{k,\frac{1}{8}}(z)\}. \qquad \ldots\ldots(28)$$

Now, it can be seen from Kummer's first formula that $z^{-3/8}M_{k,-\frac{1}{8}}(z)$ and $z^{-5/8}M_{k,\frac{1}{8}}(z)$ are both even functions of $\mu^{1/2}$, and can therefore be represented as

* See Whittaker and Watson's *Modern Analysis*, chap. 16.

absolutely convergent double power series in λ and μ. It follows from (28) that the same is true of A, and therefore also of τ, expressed by (27). The constant A_0 in (25) is given by

$$A_0 \sinh[\{\phi(\epsilon)\}^{1/2}\epsilon] = f(z_0), \qquad \ldots\ldots(29)$$

so $\tau(r)$, expressed by (25), has also a convergent expansion in powers of λ and μ. One concludes that the solution obtained of (20) is the sum of the series defined in (16).

For the practical determination of A in the limit $\epsilon = 0$, it is convenient to express $f(z)$ in (28) in terms of Whittaker functions, thus:

$$f(z) = z^{-5/8}\{A_1 W_{k,\frac{1}{8}}(z) + A_2 W_{-k,\frac{1}{8}}(-z)\} \qquad \ldots\ldots(30)$$

which has the asymptotic value

$$f(z) \sim z^{-5/8}\{A_1 z^k e^{-z/2} + A_2(-z)^{-k} e^{z/2}\}. \qquad \ldots\ldots(31)$$

Substituting this into the continuity condition of (28), one sees that A_2 must vanish in the limit $\epsilon = 0$, and

$$A \to \frac{(\frac{1}{2}\mu^{1/2})^{1/4}\Gamma(-\frac{1}{4})\Gamma(\frac{5}{8}-k)}{\Gamma(\frac{1}{4})\Gamma(\frac{3}{8}-k)} \qquad \text{as} \qquad \epsilon \to 0. \qquad \ldots\ldots(32)$$

The integral equation (21) can be solved in a similar way. Expanding κ in a series of Legendre polynomials of the cosine of the angle between $\mathbf{r}$ and $\mathbf{r}'$:

$$\kappa(\mathbf{r}, \mathbf{r}') = \sum_{l=0}^{\infty} \kappa_l(r, r') P_l(\mathbf{r} \cdot \mathbf{r}'/rr'), \qquad \ldots\ldots(33)$$

one obtains, for the determination of κ_l,

$$\kappa_l(r, r') = \frac{1}{4\pi r}\left(\frac{r'}{r}\right)^l - \frac{1}{2l+1}\left\{\int_0^r \left(\frac{r_1}{r}\right)^{l+1} + \int_r^\infty \left(\frac{r}{r_1}\right)^l\right\}\phi(r_1)\kappa_l(r_1, r')r_1\, dr_1 \qquad \ldots\ldots(34)$$

for $r \geqslant r'$. As $\kappa_l(r, r')$ is symmetrical with respect to r and r' it suffices to consider this equation. The differential equation satisfied by κ_l is

$$\frac{1}{r}\frac{\partial^2}{\partial r^2}(r\kappa_l) - \frac{l(l+1)\kappa_l}{r^2} = \phi(r)\kappa_l \qquad \ldots\ldots(35)$$

for all values except at $r = r'$, where there is a discontinuity in the first derivative. The boundary conditions are

$$4\pi r^{l+1}\kappa_l(r, r') = r'^l - \frac{1}{2l+1}\int r_1^l \kappa_l(r_1, r')\phi(r_1)\, d\mathbf{r}_1, \quad r = \infty \quad r\kappa_l = 0, \quad r = 0. \qquad \ldots(36)$$

It follows that $\kappa_l = C_l r'^{-1/2} J_{l+\frac{1}{8}}[i\{\phi(\epsilon)\}^{1/2} r] z^{-3/2} M_{k,\frac{1}{4}l+\frac{1}{8}}(z) \qquad \ldots\ldots(37)$

for $r' \leqslant \epsilon \leqslant r$ and that

$$4\pi\kappa_l \to (\tfrac{1}{2}\mu^{1/2})^{-1/4} D_l (zz')^{-3/8} M_{k,\frac{1}{4}l+\frac{1}{8}}(z) W_{k,\frac{1}{4}l+\frac{1}{8}}(z') \qquad \ldots\ldots(38)$$

for $r \geqslant r' \geqslant \epsilon \to 0$. The constant D_l is fixed by the observation that the derivative with respect to r of $r\kappa_l$ changes by $(2l+1)/(4\pi r)$ at $r = r'$; this requires

$$D_l = \Gamma(\tfrac{5}{8} + \tfrac{1}{4}l - k)\{\Gamma(\tfrac{1}{2}l + \tfrac{1}{4})\}^{-1}. \qquad \ldots\ldots(39)$$

From (36) one sees that

$$\int r_1^l \kappa(r_1, r)\phi(r_1)\, d\mathbf{r}_1 = (2l+1)\{r^l - D_l(\tfrac{1}{2}\mu^{1/2})^{l/4} z^{-3/8} W_{k,\frac{1}{4}l+\frac{1}{8}}(z)\} \qquad \ldots\ldots(40)$$

which reduces to (22) when $l = 0$.

§ 5. NUMERICAL RESULTS

The solutions of (20) and (21) obtained in the last section may now be substituted into (16), (17) and (18), thus enabling one to evaluate the coefficients in (15).

From (16) and (20) it follows that

$$R_0 = R_0' = \lim_{r=\infty} 4\pi r\{1-\tau(r)\} = -4\pi A, \qquad \ldots\ldots(41)$$

with A given by (32). Also, by substituting (33) into (18), and using (40) with $l=1$, one has

$$R_1 - R_1' = \int r\tau(r)\phi(r)\,d\mathbf{r},$$

$$\tau_1(r) = -D_1(\tfrac{1}{2}\mu^{1/2})^{1/4} z^{-3/8} W_{k,\frac{3}{8}}(z) = ar + br^{-2} + \mathrm{O}(r^{-3}), \quad \text{say.} \qquad \ldots\ldots(42)$$

Making use of the fact that τ_1 satisfies the same equation (35) as κ_1, one has

$$R_1 - R_1' = 4\pi\left[r^2\frac{d}{dr}(r\tau_1 - ar^2 - br^{-1}) - 2r(r\tau_1 - ar^2 - br^{-1})\right]_0^\infty = 12\pi b$$

$$b = \frac{(\tfrac{1}{2}\mu^{1/2})^{3/4}\Gamma(-\tfrac{7}{4})\Gamma(\tfrac{7}{8}-k)}{\Gamma(\tfrac{3}{4})\Gamma(\tfrac{1}{8}-k)}. \qquad \ldots\ldots(43)$$

To evaluate R_1, as expressed by (17), one would require more tedious calculations. However, the values already obtained suffice to determine B_+ and B_- near absolute zero. One has

$$B_+ = \tfrac{1}{2}\beta(R_0 + R_0') - \tfrac{1}{2}(\pi\beta)^{3/2}$$

$$= -\frac{4\pi(\tfrac{1}{2}\mu^{1/2})^{1/4}\Gamma(-\tfrac{1}{4})\Gamma(\tfrac{5}{8}-k)\beta}{\Gamma(\tfrac{1}{4})\Gamma(\tfrac{3}{8}-k)} - \tfrac{1}{2}(\pi\beta)^{3/2} \qquad \ldots\ldots(44)$$

$$B_- = \tfrac{1}{2}(R_1 - R_1') + \tfrac{1}{2}(\pi\beta)^{3/2}$$

$$= -\frac{6\pi(\tfrac{1}{2}\mu^{1/2})^{3/4}\Gamma(-\tfrac{7}{4})\Gamma(\tfrac{7}{8}-k)}{\Gamma(\tfrac{3}{4})\Gamma(\tfrac{1}{8}-k)} + \tfrac{1}{2}(\pi\beta)^{3/2} \qquad \ldots\ldots(45)$$

with neglect of terms of lesser orders of magnitude.

In the units chosen with $\hbar = m = 1$, $\pi\beta = 38{\cdot}04\,\text{Å}^2/T$, where T is the absolute temperature and, if Buckingham's values are accepted, $(\tfrac{1}{2}\mu^{1/2})^{1/4} = 3{\cdot}303\,\text{Å}$, $k = 0{\cdot}5432$ for helium. (These values should strictly be corrected for quantum-mechanical effects at moderately high temperatures, but the work of de Boer and Michels (1938) indicates that the error is not large.) Thus

$$B_+ = -31{\cdot}15\pi\beta - \tfrac{1}{2}(\pi\beta)^{3/2} \qquad \ldots\ldots(46)$$

$$B_- = 1125 + \tfrac{1}{2}(\pi\beta)^{3/2} \qquad \ldots\ldots(47)$$

for helium. An estimate can be made of the upper limit to the temperature range in which these formulae hold by comparing the term $31{\cdot}15\pi\beta$ in (46) with an approximation to the constant term $\tfrac{1}{2}(R_1 + R_1')$ which is the first-order correction; one finds in this way a value slightly less than 1° K. On the other hand, extended calculations of R_1, R_2, etc. could probably make the development useful up to the λ-point. At 2·61° K the uncorrected formula (46) predicts a value $-482\,\text{Å}^3$ for B_+, against an experimental value (Keesom and Walstra 1940) of $-195\,\text{Å}^3$;* it seems unlikely, therefore, that the formula could be applied above the λ-point.

* To make the necessary conversion from Amagat units, divide by the molecular number density at S.T.P.

Owing to the fact that the term proportional to β vanishes identically in the formula (45) for Fermi statistics, the effect of the molecular interactions does not set in till a temperature of a few tenths of a degree Kelvin is reached. In the case of fluids, the molecules of which obey Bose statistics, on the other hand, the deviation from ideal gas behaviour is already large at temperatures well below 0·1° K.

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The Thermal Conductivity of some Polycrystalline Solids at Low Temperatures

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Communicated by F. E. Simon; MS. received 3rd July 1952

ABSTRACT. The thermal conductivities of polycrystalline alumina, beryllia and graphite have been measured between 2° K and room temperature. The first two are dielectric solids but, even in the graphite specimens, the electronic contribution to the heat conduction is very small.

It is shown that the phonon mean free path is limited by scattering at the crystallite boundaries at sufficiently low temperatures. For sintered specimens of alumina and beryllia, in which the density is near the single crystal value, it appears that the mean free path can become greater than the crystallite size. On the other hand there is no evidence that the mean free path can increase above the crystallite size in the graphite specimens, although in this case the interpretation of the results is more difficult.

As a consequence of this restriction on the mean free path, graphite specimens with small crystallites are extremely good heat insulators at low temperatures, being better than a glass at temperatures below 5 to 10° K.

§ 1. INTRODUCTION

THE thermal conductivity of pure dielectric crystals is determined by two main factors, the interaction of the Debye waves with one another and the scattering of the waves by the boundaries of the crystal. The former give rise to a thermal resistance if the interactions are of the *Umklapp* type, and this resistance decreases with decreasing temperature; at sufficiently low

temperatures the resistance falls off exponentially. In a recent paper (Berman, Simon and Wilks 1951) this exponential variation has been discussed for the case of several single crystals. The scattering of the waves at the boundaries leads to a size-dependent conductivity at very low temperatures, an effect which was first found by de Haas and Biermasz (1938). According to Casimir's theory (1938), if boundary scattering alone were present the thermal conductivity would be proportional to the cube of the absolute temperature (as is the specific heat) and to the diameter of the crystal. De Haas and Biermasz showed that at the lowest temperatures ($\sim 2^\circ$ K) the conductivity is indeed proportional to the diameter for the thinnest crystals, but is less size-dependent for thicker crystals. None of their specimens, however, showed as strong a temperature dependence as the specific heat.

Measurements at lower temperatures have been made on potassium chrome alum by Bijl (1949) and by Garrett (1950). Between 1·5 and 3·6° K Bijl's measurements fit a $T^{2\cdot3}$ law, while Garrett found that between 0·16 and 0·29° K, within the experimental accuracy his results fitted a T^3 law.

By analogy with kinetic theory the thermal conductivity κ can be expressed in terms of the mean free path l of the phonons, or lattice waves, responsible for the conduction by the equation $\kappa = \frac{1}{3}lCv$, where C is the specific heat per unit volume and v is the phonon velocity. For boundary scattering this mean free path should be of the order of magnitude of the crystal diameter. However, in none of the above cases did the calculated mean free path reach this value. In Garrett's measurements, for example, the mean free path was only about one-twentieth of the diameter, and he attributed this to scattering at boundaries within the crystal caused by some mosaic structure.

It therefore seemed desirable to carry out further experiments on the size effect to determine whether, for any crystals, the phonon mean free path does become as great as the crystal diameter at sufficiently low temperatures. It was also interesting to see to what extent the crystallite boundaries in a microcrystalline solid limit the phonon mean free path. Since compressed crystallites of paramagnetic salts and carbon resistors are used for producing and measuring low temperatures, this investigation would also be of practical interest.

It has been possible to carry out the present investigation without going down to temperature below 2° K by taking advantage of the close connection discussed elsewhere (Berman *et al.* 1951), between the variation of thermal conductivity with temperature and the Debye characteristic temperature for specific heats Θ. The measurements have been made on crystals with high values of Θ such as diamond, graphite, alumina and beryllia so that very low relative temperatures could be obtained. As this paper is chiefly concerned with microcrystalline solids the measurements on diamond will be published separately later.

§ 2. THE EXPERIMENTS

2.1. *Apparatus*

For the measurements below 90° K the apparatus described elsewhere (Berman 1951) was used. To extend the measurements to higher temperatures a simpler apparatus was constructed in which the specimen was soldered to the base of a brass vessel which could be evacuated and immersed in a suitable fixed-temperature bath, and the temperature gradient was measured with

platinum resistance thermometers. It was satisfactory to find that the values of conductivity measured in the neighbourhood of 90° K agreed to within 2% in all cases with the values obtained with the lower temperature apparatus, in which temperatures are measured with gas thermometers.

2.2. *The Specimens*

(i) *Alumina.* The artificial sapphire single crystal measured previously (Berman 1951) was ground to a diameter of 1·5 mm from the original 3 mm. The sintered alumina was a rod of $\frac{1}{4}$ in. in diameter obtained from the Royal Aircraft Establishment, Farnborough. The density was 3·70 g/cm^3, which is 95% of the single crystal value.

(ii) *Beryllia.* This was a rod with a square cross section of side 5 mm, received from Dr. S. G. Bauer of the Atomic Energy Research Establishment. The density was 2·94 g/cm^3, which is over 97% of the single crystal value.

(iii) *Graphite.* Four types of graphite were measured. Three were artificial graphites and one was a natural graphite. The properties are shown in table 1. The artificial graphite was made by the extrusion process which produces a slight anisotropy, the crystallites tending to align with their *c*-axes perpendicular to the direction of extrusion. Cylindrical specimens were measured in which the lengths were parallel and perpendicular to the direction of extrusion. The natural graphite specimens of square cross section were cut out of a block kindly supplied by Dr. R. W. Powell of the National Physical Laboratory. The density was very close to the single crystal density of 2·25 g/cm^3 and the anisotropy was very great.

Contacts to all these specimens were made by well-fitting copper collars with a little grease inserted to take up any irregularities. It was verified that the contact thermal resistance was sufficiently small but, as an additional check, one specimen was measured with different contacts. Copper was electro-deposited on to the graphite in two rings to which the thermometers were soldered. The ends were screw-threaded and the end contacts screwed on. As the measured conductivity was not altered by this method of fixing it was not used for any other specimens.

(iv) *Carbon resistor.* This was an L.A.B. 33 ohm, $\frac{1}{2}$ watt resistor. The heater was soldered to one of the leads of the resistor and the other lead was used to attach the specimen to the apparatus. The contacts for the thermometers were made, as for the other specimens, by well-fitting copper collars.

§ 3. RESULTS AND DISCUSSION

3.1. *Alumina*

The results for the alumina specimens are shown in fig. 1. Comparing first the values for single crystals of different diameters, it can be seen that at relatively high temperatures the thermal conductivity is independent of diameter, while at the lowest temperatures, where the conductivity is nearly proportional to T^3, the conductivities are accurately proportional to the diameters. Such a general behaviour has been found on either side of the maximum for other crystals by de Haas and Biermasz. The present experiments, however, also include measurements in the neighbourhood of the maximum and extend to temperatures which are a much smaller fraction of the Debye Θ.

Table 1. Properties of the Graphite Specimens Measured

Graphite specimen	Artificial I		Artificial II		Artificial III		Natural	
Direction of length	∥ extrusion	⊥ extrusion	∥ extrusion	⊥ extrusion	∥ extrusion	⊥ extrusion	preferred direction of *c*-axis perpendicular	parallel
Crystal size ⊥ to *c*-axis (Å)	2000		1000		300		2000	
Density (g/cm³)	1·80	1·78	1·60		1·77	1·76	2·25	
Electrical resistivity (ohm cm) 290° K	$6{\cdot}0 \times 10^{-4}$	$1{\cdot}09 \times 10^{-3}$	$1{\cdot}08 \times 10^{-3}$	$1{\cdot}35 \times 10^{-3}$	$2{\cdot}33 \times 10^{-3}$	$2{\cdot}77 \times 10^{-3}$	$9{\cdot}8 \times 10^{-4}$	$4{\cdot}1 \times 10^{-3}$
90° K	$1{\cdot}09 \times 10^{-3}$	$1{\cdot}76 \times 10^{-3}$	$1{\cdot}7 \times 10^{-3}$	$2{\cdot}2 \times 10^{-3}$	$3{\cdot}01 \times 10^{-3}$	$3{\cdot}91 \times 10^{-3}$	$1{\cdot}21 \times 10^{-3}$	$5{\cdot}4 \times 10^{-3}$
20° K			$2{\cdot}3 \times 10^{-3}$	$2{\cdot}9 \times 10^{-3}$			$1{\cdot}17 \times 10^{-3}$	$5{\cdot}4 \times 10^{-3}$
4° K			$2{\cdot}3 \times 10^{-3}$	$3{\cdot}0 \times 10^{-3}$			$1{\cdot}16 \times 10^{-3}$	$5{\cdot}3 \times 10^{-3}$

Table 2. Thermal Conductivity (w/cm deg) of various Types of Solid below Room Temperature

Temperature (° K)	Room	100	50	20	10	5	2	*T* law*
Copper, Johnson Matthey spec. pure†	4	5	11·5	49	41·5	23	9·3	T
Artificial sapphire 3 mm diameter, single crystal	0·4	4	55	35	11	2·0	0·16	T^3
Artificial graphite, 300 Å crystallites	~1	0·2	0·05	0·006	0·0012	0·0003	0·00004	T^2
Quartz glass	0·01	0·006	0·003	0·0015	0·0013	0·0012	0·0005	T
Stainless steel, 18/8 Cr–Ni	0·14	0·08	0·05	0·02	0·008	0·0035	0·0012	T

* Approximate dependence on temperature at 2° K.

† Below about 20° K the values for pure copper are very sensitive to the treatment of the specimen. The values here are taken from Berman and MacDonald (1952).

Casimir (1938) calculated the heat conductivity at low temperatures by assuming that the mutual interactions between phonons can be neglected, and compared the flow of heat in a crystal with the flow of radiation along a tube with diffusely reflecting walls. Under these conditions the conductivity, if it is defined in the usual way, is shown to be proportional to the diameter of the crystal. Substituting Casimir's value of the conductivity into the equation

$$\kappa = \tfrac{1}{3} lCv \qquad \ldots\ldots(1)$$

one finds that l is almost exactly equal to the diameter for a crystal of circular cross section and to 1·1 times the length of side for a square cross section.

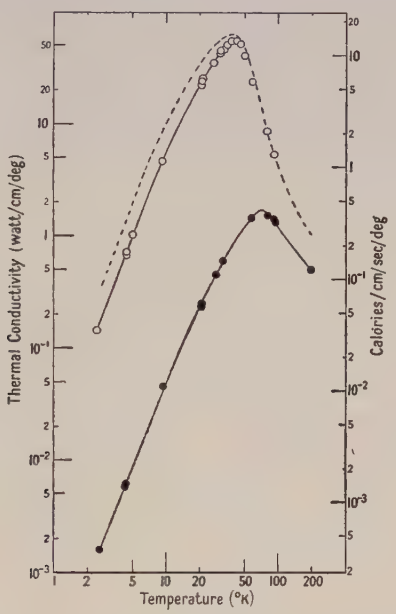

Fig. 1. Thermal conductivity of aluminium oxide.
– – – – 3 mm diameter single crystal synthetic sapphire.
○ Crystal ground to diameter of 1·5 mm.
● Sintered alumina.

The conductivity of the sapphires is found to be accurately proportional to the diameter below 10° K. Between 100 and 17° K the specific heat values of Simon and Swain (1935) and of Kerr, Johnston and Hallett (1950) can be represented by a T^3 law. If an extrapolation of the specific heat is made to lower temperatures, the phonon mean free path is calculated to be only two-thirds of the diameter at 3° K and is increasing with decreasing temperature (even at 2° K the conductivity is only proportional to $T^{2\cdot8}$). By extrapolation it is found that the mean free path would be as great as the crystal diameters at about 0·5° K.

From Klemens' theory (1951) it can be shown that near the conductivity maximum the thermal resistance of an ideal 3 mm sapphire would be about 30% greater than is given by a simple addition of resistances due to boundary scattering and *Umklapp* processes, considered as acting separately. Even so, if these were the only factors giving rise to thermal resistance, the conductivity at the maximum would be eight times as great as that measured. Some extra

cause of thermal resistance has to be assumed, which in the neighbourhood of the conductivity maximum is the chief resistance. Experiments made to study this extra resistance will be described elsewhere, but for the present it is sufficient to note that its magnitude is small enough not to enter into the discussion of microcrystalline solids which is given below.

3.2. *Sintered Alumina*

The results for sintered alumina are shown as the lowest curve in fig. 1. At the high temperature end of the range covered the conductivity is about one-half the single crystal value. At the lowest temperatures the sintered rod has a conductivity one-hundredth of that of the 1·5 mm diameter crystal, but this fraction increases slightly as the temperature decreases.

It is reasonable to assume that the reduction by a factor two at high temperatures can be accounted for by the geometry of the crystallites. Even if there is no actual contact resistance between separate crystallites, if they are not in contact over their whole surface the measured conductivity of the rod will be less than the true conductivity of the material of the crystallites. This reduction factor should be the same at all temperatures. The reduction in conductivity at the lowest temperatures due to the size of the crystallites is therefore mainly due to scattering of phonons at the boundaries of the crystallites, the process which also limits the thermal conductivity of single crystals at sufficiently low temperatures. As the ratio of the conductivity to specific heat increases as the temperature falls the phonon mean free path must be increasing with decreasing temperature. As the specimen was sintered the crystallite boundaries are slightly less definite than in a single crystal, and the phonon mean free path is able to increase slightly. The mean free path calculated from the conductivity at the lowest temperatures will therefore be greater than the crystallite size. If it is assumed that the reduction factor of two due to the geometry of the crystallites operates at all temperatures, then at the lowest temperatures an extra factor of 50 compared with the 1·5 mm crystal must be due to boundary scattering in the crystallites. The phonon mean free path in the 1·5 mm crystal at 3° K is 1·0 mm, so that the crystallites must be less than 20 μ in size.

It is also possible to obtain a rough estimate of the crystallite size from the position and value of the conductivity maximum. The variation of phonon mean free path for sapphire at temperatures above that of the conductivity maximum was shown in a previous paper (Berman *et al.* 1951). For the sintered rod the maximum is seen to occur at about 75° K, and at this temperature the mean free path due to *Umklapp* processes is 2·7 μ. The resistance of $\frac{1}{80}$ watt^{-1} calculated to be present at the single crystal maximum and ascribed to defects will be negligible compared with the resistances at 75° K due to *Umklapp* processes and to boundary scattering for the sintered rod, so that these processes should be the only two which need be considered. Simple addition of these resistances gives a resistance slightly less than is given by the more refined treatment of Klemens, but gives about the right position of the maximum. The maximum will occur when the mean free path due to *Umklapp* processes is of the order of magnitude of the crystallites, which must therefore be about 3 μ.

If at 75° K only boundary scattering and *Umklapp* processes are important, the crystallite size can be estimated from the value of the conductivity. The

measured conductivity is 1·6 watt units, so that the conductivity of the individual crystallites must be about 3·2 watts. The phonon mean free path, calculated from this conductivity according to eqn. (1), is about 1 μ. As has been mentioned above, this mean free path is slightly less than the value given by simple addition of resistances, and this, in turn, must be half the mean free path due to each scattering process acting separately. The mean free path due to boundary scattering at the conductivity maximum is therefore 2 to 3 μ, which gives the order of size of the crystallites.

The conductivity at the lowest temperatures therefore suggests a mean crystallite size less than 20 μ, while the position and value of the conductivity maximum gives a value of 2 to 3 μ. This order of size agrees quite well with direct measurement on a photomicrograph, which showed grains ranging in size from about 5 to 30 μ.

A very exact comparison between the single crystals and the polycrystalline alumina is not possible as the crystal forms may be different, and this would affect slightly some of the parameters determining thermal conductivity.

3.3. *Sintered Beryllia*

The conductivity of the sintered beryllia specimen is shown in fig. 2. Below about 30° K the conductivity is proportional to $T^{2\cdot5}$, which indicates that as the temperature decreases the phonon mean free path increases, in the same way as for sintered alumina. Preliminary measurements (Berman and Poulter, to be published) show that in this temperature region the specific heat per mean g atom is three-quarters of that of alumina. If the specific heat per mean gram atom is expressed in terms of a Θ value and the phonon velocity calculated from this value of Θ, then it is found that for the same size crystallites the conductivity of both beryllia and alumina should be very similar. A photomicrograph showed crystallites with dimensions between 10 and 40 μ, which is similar to the range of size found in the sintered alumina specimen. The conductivity at low temperatures, however, is about 40% less but, in view of the difficulty of making any exact calculations of the conductivity, this is not incompatible with the discussion of the size dependence of conductivity which has been given.

3.4. *Graphite*

The experimental results are shown in fig. 3.

Although the general interpretation of the graphite results is similar to that of the sintered alumina and beryllia, the details are more complicated for several reasons: (i) results for single crystals are not available, (ii) there is a small electronic contribution to the total heat conductivity, and (iii) graphite is very anisotropic and the specimens measured had different degrees of orientation of the crystallites.

At the lowest temperatures the heat conductivity of the artificial graphite is roughly proportional to the estimated crystallite size. An exact comparison cannot be made because of the differing densities. Kinchin (to be published) has measured the electrical conductivity of many artificial graphites and has found that for specimens with the same crystallite size but with different densities the electrical conductivity is proportional to a power of the density of the order of 4. The only indication of the effect of density in the present

experiments is given by a comparison of the natural graphite specimen and the artificial graphite I, in both of which the crystallite size is about 2000 Å. The fourth powers of the densities are in the ratio of 2·5 : 1, while the conductivities at the lowest temperatures are in the ratio of 4 : 1, so that in this case the relation does not seem to hold.

The electronic contribution to the heat conductivity can be estimated from the electrical conductivity. If the Wiedemann–Franz–Lorenz law is assumed to hold, then for the artificial graphite specimens the electronic contribution

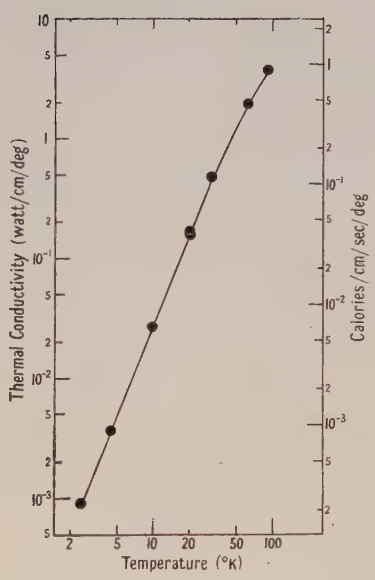

Fig. 2. Thermal conductivity of sintered beryiila.

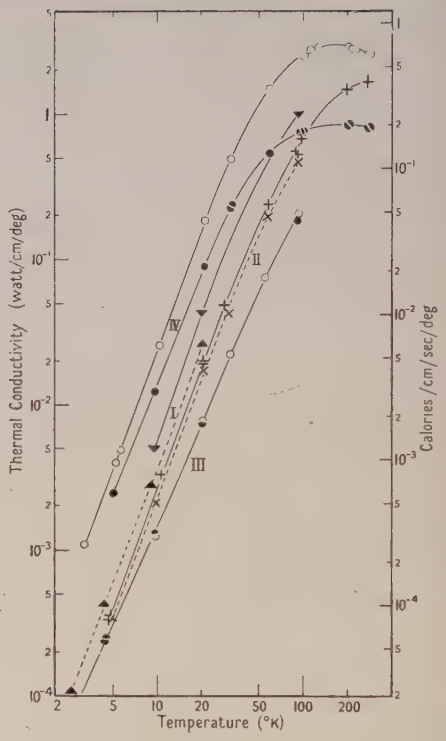

Fig. 3. Thermal conductivity of graphite.
I, II and III Artificial graphites
▼, +, ●, ∥ extrusion. ▲, ×, ○, ⊥ extrusion.
IV. Natural graphite
○, ⊥ preferred direction of C axis
●, ∥ preferred direction of C axis

increases from less than $\frac{1}{2}\%$ at room temperature to about 10% at the lowest temperatures and for the natural graphite the contribution increases from $\frac{1}{4}\%$ to 3% over the same temperature range.

The specific heat of graphite has been measured recently down to 8° K (Bergenlid and Hill, to be published). Below 20° K the specific heat is proportional to a power of the temperature of the order of 2·2. This deviation from the usual T^3 law can be ascribed to the two-dimensional layer structure of graphite, the binding between the layers of carbon hexagons being very small (Krumhansl, to be published).

It is only for the specimen with the smallest crystallite size and in which, judging by the equality of the conductivities measured in the two perpendicular

directions, the orientation of the crystallites is most random, that the conductivity varies as a power of the temperature as low as 2·2. For the other specimens, including the natural graphite, in which the orientation of the crystallites is quite great, the conductivity at low temperatures is proportional to a power of the temperature between 2·5 and 2·7.

As it is improbable that the phonon mean free path is actually decreasing with decreasing temperature, the fact that the thermal conductivity decreases more rapidly than the specific heat must either be due to an increasing contact resistance between the crystallites or must in some way be associated with the very great anisotropy. Until there is a detailed theory of the heat conductivity of such an anisotropic solid, or until measurements have been made on a single crystal of graphite, the explanation of this effect is uncertain.

It is, however, interesting to make an estimate of the phonon mean free path in the specimen with the smallest crystallite size. Using the measured values of the specific heat and of the thermal conductivity at 10° K, and taking the single crystal density as 2·25 g/cm^3, the mean free path in the crystallites is $1{\cdot}3\,z/v$ cm, where v is the mean phonon velocity and z is the factor by which the conductivity is reduced on account of the crystallite packing. It is not certain what velocity should be taken, but taking a value of 10^6 cm/sec, corresponding to the tight binding in the planes, the phonon mean free path is of the order of $100z$ Å. Taking the reduction factor as equal to the fourth power of the ratio of the single crystal density to that of the polycrystalline specimen, z is about 4. From the thermal conductivity the phonon mean free path is therefore estimated to be about 500 Å, which is in reasonable agreement with the value of the crystallite size measured by x-ray methods.

The size-dependence of conductivity can also be seen by comparing the conductivity of this specimen with that of the carbon measured by Buerschaper (1944). At 82° K the conductivity of the carbon was one-tenth of that of the graphite of smallest crystallite size. As stated by Mizushima and Okada (1951), the crystallite size determined for many kinds of amorphous carbon by x-ray methods is several tens of ångströms. The conductivity ratio is therefore of the same order of magnitude as the ratio of crystallite sizes. The density of Buerschaper's carbon is not stated, so that the extent of this agreement is not certain.

Mizushima and Okada (1951) and Mrozowski (1952) have compared the heat conduction in microcrystalline graphite to that in a glass in which at high temperatures (>200° K) the mean free path is constant and of the order of size of a 'unit cell'; this for quartz glass, for example, is the size of the SiO_4 tetrahedra ($\sim$7 Å). The comparison does not seem justifiable, however, since in a glass the mean free path increases at lower temperatures, where the dominant phonons correspond to waves with wavelengths larger than this 'unit cell' and the conductivity falls less rapidly than the specific heat (Berman 1949). Below about 20° K the mean free path is, in fact, inversely proportional to the square of the temperature, and from Klemens' (1951) theory it can be deduced that at about $\frac{1}{20}$° K the phonon mean free path would be several millimetres ($\sim 10^7$ 'unit cells') and therefore of the same order of magnitude as the specimen itself. For the various graphites and carbons which have been measured there is no experimental evidence that the phonon mean free path increases above the crystallite size with decreasing temperature. Even for

polycrystalline materials, such as sintered alumina and beryllia, which have a density nearly equal to the single crystal value, the conductivity only falls slightly less rapidly than the specific heat, indicating that the phonon mean free path increases slowly with decreasing temperature.

The large increase in the phonon mean free path with decreasing temperature in a glass has an interesting consequence which has already been noted (Berman 1949). Even at the lowest temperatures at which measurements have been made, the mean free path in the graphite with smallest crystallites does not seem to be larger than a few hundred ångströms, and this accounts for the low conductivity. At high temperatures, although the mean free path is still the same, since the specific heat is larger, the conductivity increases and at room temperature specimens of graphite can have heat conductivities similar to those of the best metals (see, for example, Powell and Schofield 1939). For a

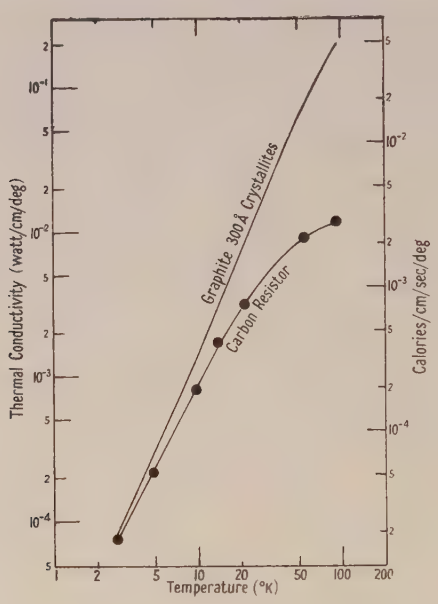

Fig. 4. Thermal conductivity of carbon resistor and artificial graphite III.

glass, however, at high temperatures the mean free path is only a few ångströms, with a correspondingly small conductivity. The conductivity does not fall with temperature as rapidly as the specific heat, and as a consequence a glass is a better conductor than a microcrystalline graphite below 5 to 10° K, and at 2° K the ratio of the conductivity of quartz glass to that of graphite III is 12. On the assumption that both conductivities can be extrapolated to lower temperatures this ratio would be 250 at 0·1° K.

3.5. *Carbon Resistor*

The thermal conductivity of the carbon resistor is shown in fig. 4 (together with that of the graphite with smallest crystallite size). From the measured electrical conductivity the electronic contribution to the heat conductivity is of the order of 10^{-3} % of the total at all temperatures below 100° K. At all

temperatures the heat conductivity is smaller than that of the graphite with the smallest crystallite size. At 100° K the ratio is 20, but decreases to 1·1 at 2·7° K. This is the variation which would be expected from a mixture of graphite of small crystallite size bonded together by some material of low heat conductivity. At the higher temperatures the graphite conducts quite well but the overall conductivity is restricted by the bonding material, and at 100° K the conductivity is only about double that of a glass and at 82° K is 25% smaller than the value for carbon given by Buerschaper. At the lowest temperatures the graphite is a poor conductor, but there is sufficient conduction through the bonding material for the overall conductivity to be greater than that corresponding to the small crystallite size of the graphite.

If carbon resistors are used as thermometers at low temperatures it is important to realize that a heat flow of only 8 erg/sec through the particular resistor measured in these experiments would, at 2° K, produce a temperature gradient in the resistor of $\frac{1}{10}$°.

§ 4. SUMMARY

It has been shown that, to within a small degree of uncertainty, the thermal conductivity of polycrystalline dielectric solids can be estimated from the conductivity of the single crystal if the crystallite size is known. At temperatures low enough for the thermal conductivity to be limited only by scattering of the phonons at the boundaries it is possible to make the estimation without a knowledge of the single crystal values if the specific heat is known. It is shown that the assumption that the phonon mean free path under these conditions is approximately equal to the crystallite size is justified even for sintered solids in which the density is very close to the single crystal value.

At high temperatures the thermal conductivity of pure dielectric solids is limited by *Umklapp* processes, so that the effect of grain size is only a secondary effect connected with the area of contact of the crystallites and the conductivity of microcrystalline materials can be little less than the single crystal value.

The conductivity of microcrystalline solids is, therefore, much more temperature dependent than that of single crystals and, as is shown in table 2, varies from values characteristic of metals, at high temperatures, to values much less than those of a glass, at low temperatures.

The table shows how the thermal conductivity varies with temperature for several types of solid and gives an indication of the kind of substance which should be employed at various temperatures in cases where the thermal and electrical conductivities are of importance.

ACKNOWLEDGMENTS

I would like to thank Mr. T. M. Fry of the Atomic Energy Research Establishment for providing the graphite specimens and for informing me of the work of G. H. Kinchin prior to its publication. I am also very grateful to Dr. W. W. Tyler of General Electric Co., Schenectady, for informing me of his experimental work on the conductivity of graphite and of the work of J. A. Krumhansl on the theory of its specific heat prior to publication.

I should like to thank Professor F. E. Simon for his continued interest in this work.

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A Note on Oriented Crystalline Overgrowth

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MS. received 24th July 1952

ABSTRACT. A brief review is given of experimental results on the oriented overgrowth of very thin deposits of alkali halides evaporated on to cleavage surfaces. The theoretical implications of some of these results are considered.

§ 1. INTRODUCTION

Recent experimental work by Schulz (1951 a, b) has provided much-needed evidence on the structure of very thin layers of deposits evaporated on to cleavage faces of crystals. In the extreme case the layers (alkali halides) had a mean thickness of 10 Å and less; it therefore appears that the initial stages of growth have been studied to the limit possible with the electron diffraction technique. In one of the sets of experiments mica substrates were used, in the other cleavage faces of alkali halides.

In the latter experiments oriented overgrowth was always obtained whatever the value of the misfit (or mismatch in the terminology used by Schulz), i.e. the relative difference in spacing of the two crystals in their normal state. This experimental result is particularly interesting in view of the theory of Frank and van der Merwe (1949) which predicted a critical value of the misfit above which oriented overgrowth should change into a randomly arranged overgrowth. The experiments show that the theory is incorrect in this respect. Further, according to the theory, the initial oriented layer should be strained to fit the substrate, i.e. should be pseudomorphic. The experimental results on deposits evaporated on to the cleavage faces of alkali halides show that where a monolayer is formed it has the normal lattice spacing (this point is not specifically emphasized by Schulz, and has been clarified in correspondence with the author). This result is in disagreement with the Frank–van der Merwe theory, which has also been criticized on theoretical grounds when applied to certain specific cases (Smollett and Blackman 1951, Blackman 1951). The

monolayers on alkali halide substrates are formed over a range of misfits from -20% to $+20\%$; outside this region oriented three-dimensional nuclei were found in the initial stages.

The experimental results on mica substrates present a somewhat different picture, one aspect of which will be discussed theoretically. Evaporated layers of alkali halides ranging in spacing from 7·32 Å (RbI) to 6·58 Å (KBr) orientate in the initial stage as a 'monolayer' with an arrangement and spacing which coincides with the sites of the potassium ions in the cleavage plane of muscovite, i.e. the initial layer is pseudomorphic. The orientation is such that (111) planes of the alkali halide are parallel to the surface of the substrate. Hence the initial 'monolayer' must contain two charges planes of ions. This type of monolayer cannot be expected to have the properties of a cubic lattice, in fact one needs three such layers to form the first cell of the three-dimensional lattice. The orientation taken up by such a monolayer should therefore be considered in relation to the monolayer in its 'free' state, i.e. without the substrate.

It will be shown (§2) that the state of lowest energy (if the hexagonal symmetry is maintained) is one in which the positive and negative ions lie in the same plane, which is then neutral. The pseudomorphic layer which is found experimentally should be considered from the point of view of this neutral plane.

Beyond the monolayer range the alkali halides form initially oriented three-dimensional nuclei on mica, as with the alkali halide substrates.

§2. THE POTENTIAL ENERGY OF THE MONOLAYER

The monolayer on mica consists of two oppositely charged planes with an arrangement of ions as in the (111) planes of the alkali halide. The distance between the planes is treated as a parameter in the following calculations. The potential energy of the layer will be proportional to the potential energy of a representative ion in the field of all the others, if the repulsive forces are taken to be the same for the two types of ions. The multiplying factor will be N, where N is the number of ions of any one type.

The electrostatic part of the potential energy will be considered first. The two-dimensional lattice formed by the ions of the same sign is hexagonal, the lattice vectors being $\mathbf{a}_1$ and $\mathbf{a}_2$ respectively, where $a_1 = a_2 = a$ and the angle between the vectors is 120°. The distance between the two layers is a_3.

The lattice distance $r_{11}^{l_1 l_2}$ between ions of type one can then be expressed as

$$(r_{11}^{l_1 l_2})^2 = a^2(l_1^2 + l_2^2 - l_1 l_2). \qquad \ldots\ldots(1)$$

The corresponding distance between ions of mixed type can then be written as

$$\begin{aligned}(r_{12}^{l_1 l_2})^2 &= (l_1 + \tfrac{1}{3})^2 a_1^2 + (l_2 + \tfrac{2}{3})^2 a_2^2 + 2(l_1 + \tfrac{1}{3})(l_2 + \tfrac{2}{3})(\bar{\mathbf{a}}_1 . \bar{\mathbf{a}}_2) + a_3^2 \\ &= a^2(l_1^2 + l_2^2 - l_1 l_2 + l_2 + \tfrac{1}{3} + f), \qquad \ldots\ldots(2)\end{aligned}$$

where $a_3^2 = f a^2$. Here the ions of type two are also arranged as in the (111) alkali halide planes, but the distance between the planes, taken as a_3, is treated as a parameter, fixed by the value of $f^{1/2}$.

The electrostatic potential energy of an ion can then be written as

$$E = \frac{e^2}{a} \Sigma \{[\phi(l)]^{-1/2} - [\phi(l+g) + f]^{-1/2}\}, \qquad \ldots\ldots(3)$$

where

$$\phi(l) \equiv l_1^2 + l_2^2 - l_1 l_2, \quad \phi(l+g) \equiv l_1^2 + l_2^2 - l_1 l_2 + l_2 + \tfrac{1}{3} \qquad \ldots\ldots(4)$$

in the notation used by Epstein (1903). The Epstein method, first introduced by Emersleben (1923) into lattice theory, will be used in handling the above sum. The general formulae given by Epstein are, however, applicable only to a complete lattice, and not in the above case. The mathematical method can still be used, and leads to extra terms in the usual formulae. The result is that E can be expressed in the form

$$E=\frac{e^2}{a}\Big[-2+\int_1^\infty dz\, z^{-1/2}\,\Sigma\,\{\exp[-\pi z\phi(l)]-\exp[-\pi z\phi(l+g)-\pi zf]\}$$
$$+\frac{2}{\sqrt{3}}\int_0^1 dz\, z^{-3/2}\{1-\exp(-\pi fz)\}$$
$$+\frac{2}{\sqrt{3}}\int_1^\infty dz\, z^{-1/2}\sum_{m_1m_2}{}'\exp[-\pi z\Phi(m)]$$
$$-\frac{2}{\sqrt{3}}\int_1^\infty dz\, z^{-1/2}\exp[-\pi f/z]\sum_{m_1m_2}{}'\exp[-\pi z\Phi(m)]\exp[-\tfrac{2}{3}\pi i(m_1+2m_2)]\Big] \quad \ldots\ldots(5)$$

where
$$\Phi(m)\equiv\tfrac{4}{3}(m_1^2+m_2^2+m_1m_2) \quad \ldots\ldots(6)$$

and the zero term is to be omitted in the summations over m_1, m_2. The expressions for ϕ have been given in (4).

It will be noted that if one puts $f=0$, the expression (5) reduces to a standard form of the Epstein type, which holds for the particular two-dimensional lattice obtained by this operation. With one exception all the above integrals can be expressed in terms of known functions (exponential or error functions). The last integral has to be handled in part numerically, but it can be arranged in such a way that its contribution can be evaluated with sufficient accuracy without difficulty. A representative set of the results of the calculations of E for various values of f in units of $-e^2/a$ is

f	0·00	0·02	0·04	0·06	0·08	0·10
E	2·6710	2·4466	2·2350	2·0348	1·8446	1·6633

That part of the potential energy due to the repulsive forces has been evaluated using a simple power law of the form br^{-n}. Two cases were chosen: $n=10$ and $n=7$ respectively. The constant b is expressed, where convenient, in terms of the distance r_0 between the positive and negative ions along the cube axis of the three-dimensional crystal; for instance, for $n=10$, $b=1{\cdot}747e^2r_0^9/60{\cdot}36$.

The value of the repulsive part of the potential energy R can be written (for $n=10$) in the form

$$R=6bS/a^{10}, \quad \ldots\ldots(7)$$

where a is the lattice spacing defined above. S depends only on the value of f, and is evaluated numerically for various values of f. For each of these values the magnitude of a/r_0, which makes the total potential energy $U=E+R$ a minimum is first found, and this minimum value of U then follows. This procedure has been carried through for both values of n; representative energy values $U(n)$ in units of $-e^2/r_0$ and magnitudes of a/r_0 are:

f	0·00	0·02	0·04	0·06	0·08	0·10
$U(10)$	1·3785	1·2914	1·2042	1·1168	1·0292	0·9417
$U(7)$	1·3358	1·2467	1·1577	1·0689	0·9805	0·8923
$a/r_0(10)$	1·744	1·705	1·671	1·640	1·613	1·590
$a/r_0(7)$	1·714	1·682	1·655	1·632	1·612	1·598

The value of f in the three-dimensional crystal is one-sixth. In the process of decreasing f the lattice spacing a increases and for $f=0$, the value of a becomes $1{\cdot}233(2)^{1/2}r_0$ for $n=10$, and $1{\cdot}212(2)^{1/2}r_0$ for $n=7$. In the three-dimensional crystal $a=(2)^{1/2}r_0$.

§ 3. DISCUSSION

It will be seen from the above table that the minimum value of the potential energy of the 'free' monolayer is obtained when $f=0$, i.e. when a neutral plane is formed. It seems therefore necessary to consider misfits in terms of the spacing of this neutral plane rather than in terms of that of the three-dimensional crystal. Defining the misfit as $(a-a')/a'$, where a' is the spacing of the mica and a that of the neutral plane, it is found that the pseudomorphic region is characterized by misfits ranging from 20% (for RbI) to 8% (for KBr).

It is noteworthy that small misfits (less than 7%) are not found in the pseudomorphic region—three-dimensional nuclei being formed in preference. It therefore appears that the base differentiates strongly between the positive and the negative ions, and also that the monolayer contains well-separated planes of ions. A lower limit to this separation can be obtained if one considers the 'free' double layer (with the appropriate value of the a spacing) and ignores the effect of the base. This limit follows from the calculation in § 2, which gives the value of a/r_0 for a definite value of f in a state of minimum energy. The distance between the planes is found to be 2·1 Å for RbI and 1·2 Å for KBr with intermediate values for alkali halides of intermediate spacing.

That the initial deposited layer can be either an oriented pseudomorphic monolayer or an oriented three-dimensional nucleus is not in accord with the Frank–van der Merwe theory, which predicts either an oriented pseudomorphic layer (associated with small misfits) or a non-oriented 'dislocated' layer of normal average spacing. The disagreement is further emphasized by the absence of a region of small misfit associated with pseudomorphic monolayers.

The discussion given so far applies in the main to monolayers on mica. It is also found experimentally (Schulz 1951 a) that the normal crystal structure is obtained when the thickness of the deposit is increased. This will also follow from the above calculations, as both the distance between planes and the spacing in a plane will tend to their value in the three-dimensional crystal as the number of layers is increased. There must, however, be a rearrangement at some stage, the details of which will need further calculation. Similarly, the behaviour of alkali halides on mica when their spacing is less than 6·58 Å, i.e. the formation of three-dimensional nuclei, still awaits consideration. It is evident that much more theoretical work will be needed before the initial stages of oriented overgrowth are understood, even in those cases for which experimental data are available.

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The Velocity of Second Sound in Various Media

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Communicated by H. N. V. Temperley ; MS. received 20th February 1952

ABSTRACT. On the assumption that second sound can be propagated through media other than helium II, its velocity is calculated for non-ideal gases, insulators, conductors, superconductors and ferromagnets.

§ 1. INTRODUCTION

In helium II two types of wave propagation are possible: ordinary 'first sound', in which pressure and density fluctuations predominate, and 'second sound', in which the fluctuations are primarily in temperature and entropy. According to the present theory of second sound (for review, see Dingle 1952), its velocity c_2 is given by the relation

$$c_2^2 = \frac{TS^2}{C\rho_n}\left(1-\frac{\rho_n}{\rho}\right) = -\frac{\partial T}{\rho_n \partial(1/S)}\left(1-\frac{\rho_n}{\rho}\right), \qquad \ldots\ldots(1.1)$$

where T is the absolute temperature, ρ the density of the medium, S and C respectively the entropy and specific heat per unit volume, and ρ_n the effective mass-density of the excitations which contribute to the entropy. In terms of the energy spectrum $E=E(p)$ relating the energy E of the excitations to their momentum p,

$$C=(\partial U/\partial T)_V, \qquad S=\int_0^T C\,dT/T, \qquad \ldots\ldots(1.2)$$

where U is the internal energy per unit volume given by

$$U=\frac{4\pi}{h^3}\int \frac{Ep^2\,dp}{\xi \exp(E/\boldsymbol{k}T)\mp 1}. \qquad \ldots\ldots(1.3)$$

The effective mass-density of the excitations is

$$\rho_n = -\frac{4\pi}{3h^3}\int p^4 \frac{\partial}{\partial E}\frac{1}{\xi \exp(E/\boldsymbol{k}T)\mp 1}. \qquad \ldots\ldots(1.4)$$

For free particles of mass m, $E(p)=p^2/2m$, and (1.4) simply yields the expected value $\rho_n=Nm$, where N is the number of particles per unit volume.

The theoretical conditions which would have to be satisfied before second sound could be propagated through a medium are:

(i) ρ_n/ρ must be less than unity, for otherwise (1.1) would predict an imaginary value for c_2. The physical meaning of this result is that second sound arises through relative motion between the excitations and the configuration of the ground state of the assembly, and can only occur if the excitations are the more easily movable of the two, i.e. if they possess the lower effective mass.

(ii) The coefficient of thermal expansion must be small: if it is not, the temperature and entropy fluctuations characteristic of second sound are accompanied by considerable density and pressure fluctuations, and conversely

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the density and pressure fluctuations characteristic of first sound are accompanied by considerable fluctuations in temperature and entropy, with the result that the two sounds could not be excited separately. For calculations of the coupling see Lifshitz (1944), Peshkov (1948) and Dingle (1950).

(iii) The mean free path of the excitations must be less than the wavelength (velocity frequency) of second sound it is proposed to excite (also noted by London (1951) and Ward and Wilks (1952)). This condition arises on account of the assumption in the theory that local equilibrium is attained within a distance much smaller than that in which there is any appreciable fractional change in the temperature, entropy, etc. Otherwise, of course, the use of the *equilibrium* distribution function $1/(\xi \exp(E/\boldsymbol{k}T) \mp 1)$ would not be justified.

(iv) Collisions between excitations must satisfy the laws of conservation of energy and momentum, for collisions of the 'Umklapp'* type would probably lead to a diffusion equation rather than to a wave equation (Ward and Wilks 1951, 1952).

(v) Collisions between excitations must not lead to any fundamental change in the energy spectrum. For example, in an assembly of Bloch spin waves in a ferromagnet (Bloch 1930) this condition would be violated except at very low temperatures, because the simple addition of energies of separate spin-waves is only justified if the probabilities of reversed spins being on neighbouring atoms is minute.

(vi) Irreversible processes such as viscosity, thermal conduction and relaxation must not lead to a severe attenuation. For discussion, formulae and references see review (Dingle 1952, § 6).

In spite of our limited knowledge of the nature and properties of excitations in helium II it is possible to see some reasons why this substance should be so favourable as a medium for the propagation of second sound. For according to the theory of helium II as a condensed Bose–Einstein assembly, ρ_n/ρ *is* less than unity, thus satisfying (i), but not so much less as to give rise to difficulty. (If ρ_n/ρ is extremely small, the drift velocity of the excitations becomes extremely large even for second sound of small amplitude; this leads to irreversible processes and hence severe attenuation.) As for condition (ii), the coefficient of thermal expansion of helium II is very small ($\sim -3 \times 10^{-3}$ deg^{-1}). Collisions between excitations in which their momentum is not conserved are probably relatively unlikely at helium temperatures,† so that condition (iv) is likely to be satisfied. Finally, with reference to (vi), the viscosity and thermal conduction of liquid helium are quite small.

In this paper we attempt to estimate the velocity of second sound to be expected in non-ideal gases, insulators, conductors, superconductors and ferromagnets, assuming that experimental conditions can in fact be so chosen that none of the theoretical limitations (i) to (vi) are transcended.

§ 2. VELOCITY IN NON-IDEAL GASES

In an *ideal* gas composed of particles of mass m, $E(p) = p^2/2m$, and (1.4) gives $\rho_n = Nm$, where N is the number of atoms per unit volume. Hence $\rho_n = \rho$, and there can be no second sound.

* Roughly speaking, Bragg reflection of phonons by the lattice ; in this process phonon momentum is not conserved.

† For example, ' Umklapp ' processes are possible only if phonons have a certain minimum energy, and the probability of their possessing this is less the lower the temperature.

When the gas is not ideal, second-sound propagation may become possible, the forces called into play being the same as those responsible for thermal diffusion. If the interatomic forces may be adequately represented by a self-consistent field, we may take $E(p)=Ap^{1/r}$, where A and r are parameters (Dingle 1952, § 3.22). Then, on classical statistics,

$$\frac{\rho_n}{\rho}=\frac{(\boldsymbol{k}T)^{2r-1}\Gamma(5r)}{3mA^{2r}\Gamma(3r)}, \qquad \ldots\ldots(2.1)$$

which may be less than unity, its value for an ideal gas in which $r=\frac{1}{2}$ and $A=1/2m$. On the same model

$$C=3N\boldsymbol{k}r, \qquad S=3N\boldsymbol{k}r(\ln T)+\text{constant}. \qquad \ldots\ldots(2.2)$$

For an actual gas ρ_n/ρ will be very close to unity, and the situation will therefore be similar to that of helium II at temperatures immediately below the lambda-point—i.e. the velocity will be small, and the attenuation due to viscosity and thermal conduction large. It must also be noted that condition (ii), that the coefficient of thermal expansion should be small, is not satisfied, and as a result there is strong coupling between the sounds. As far as the velocity of second sound is concerned the quantity c_2 given by eqn. (1.1) should be multiplied by approximately $\gamma^{-1/2}=(C_p/C_v)^{-1/2}$.

The values of ρ_n/ρ and c_2 in a condensed Bose–Einstein gas have already been fully discussed elsewhere (Dingle 1952, § 4.32).

§ 3. VELOCITY IN INSULATORS

Peshkov (1947) and Ward and Wilks (1951) have explicitly referred to the possibility that second sound might be propagated through solids. For an insulating solid at low temperatures the only carriers of entropy are phonons (Debye waves). Taking c_l and c_t to be respectively the velocities of longitudinal and transverse waves of ordinary first sound, and assuming the temperature to be considerably lower than the Debye characteristic value, (1.2), (1.3) and (1.4) give

$$S=\frac{16\pi^5\boldsymbol{k}^4T^3}{45\hbar^3}\left(\frac{1}{c_l^3}+\frac{2}{c_t^3}\right), \qquad \ldots\ldots(3.1)$$

$$\rho_n=\frac{16\pi^5\boldsymbol{k}^4T^4}{45\hbar^3}\left(\frac{1}{c_l^5}+\frac{2}{c_t^5}\right), \qquad \ldots\ldots(3.2)$$

so that at very low temperatures, where $\rho_n/\rho\ll 1$, the velocity of second sound is given by (1.1) as

$$c_2^2=\left(\frac{1}{c_l^3}+\frac{2}{c_t^3}\right)\Big/3\left(\frac{1}{c_l^5}+\frac{2}{c_t^5}\right). \qquad \ldots\ldots(3.3)$$

(This formula, a generalization of one due to Landau (1941), was given by Temperley (1952) as applying to helium II, but other authors have assumed that transverse waves cannot be propagated through this liquid.)

§ 4. VELOCITY IN CONDUCTORS

We consider first a degenerate assembly of free electrons in isolation. The entropy and specific heat per unit volume are

$$S=C=\gamma T, \qquad \ldots\ldots(4.1)$$

where γ is the Sommerfeld coefficient, $\gamma = \frac{1}{2}\pi^2 N\boldsymbol{k}/T_0$, N being the number of electrons per unit volume and T_0 the degeneracy temperature. Since the electrons are assumed to be free, we must take

$$\rho_n = Nm \qquad \ldots\ldots(4.2)$$

so that $\rho_n = \rho_{\text{electrons}}$ and second-sound propagation is impossible.

The state of affairs in an actual metal is more complicated. We make the usual approximations of the theory of metals, and thus assume that the ions form a nearly rigid lattice through which quasi-free electrons, of effective mass m, may move. Except at absolute zero, the mean free paths of both phonons (vibrations of the lattice) and electrons will be finite, and it should be possible to satisfy condition (iii) for both. Under these circumstances it seems reasonable to add electronic and phonon contributions, and take

$$S = S_{\text{phonons}} + S_{\text{electrons}}, \qquad \rho_n = (\rho_n)_{\text{phonons}} + (\rho_n)_{\text{electrons}}. \qquad \ldots\ldots(4.3)$$

At reasonably high temperatures ($T \gtrsim 100^\circ$ K for most metals) the electronic contributions will all be negligible, and c_2 will be given by (3.3), as for insulators. At lower temperatures (100° K $\gtrsim T \gtrsim 2^\circ$ K) the lattice vibrations provide the dominant part of the entropy and specific heat, but the effective mass is mainly electronic in origin; in this region

$$c_2^2 \simeq \frac{16\pi^5 \boldsymbol{k}^4 T^4}{135 h^3 N m}\left(\frac{1}{c_l^3} + \frac{2}{c_t^3}\right), \qquad \ldots\ldots(4.4)$$

so that the velocity falls rapidly as the temperature is lowered, ultimately attaining a value of the order of a few metres per second. At extremely low temperatures ($T \lesssim 2^\circ$ K) the lattice contributions are all negligible, and the velocity would be

$$c_2 = T(\gamma/Nm)^{1/2} = \pi T(\boldsymbol{k}/2mT_0)^{1/2} = \pi \boldsymbol{k} T/p_0, \qquad \ldots\ldots(4.5)$$

where p_0 is the momentum at the Fermi surface. Numerically (4.5) yields the result

$$c_2 = 8{\cdot}66 \times 10^3 T/\sqrt{T_0} \text{ m/s}, \qquad \ldots\ldots(4.6)$$

so that for most metals c_2 would be about $40T$ m/s at extremely low temperatures. These figures lead us to expect a minimum in the velocity–temperature curve just above about 2° K, and a maximum just below (fig. 1).

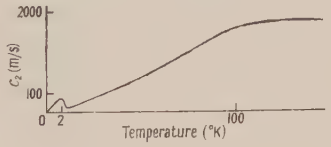

Fig. 1. Velocity of second sound in a conductor.

So far, it has been assumed that the mean free paths of both phonons and electrons are smaller than the wavelength of second sound, i.e. that the frequency is low. At higher frequencies the electrons will not remain in equilibrium with the phonons, and second sound will travel through the lattice only, its velocity being given by (3.3), as for insulators. In the intermediate frequency region there will be a relaxation effect between the electron and lattice systems which will lead to an attenuation additional to that expected from ordinary irreversible processes, thermal conduction and ohmic resistance being the most serious of these for a metal.

§ 5. VELOCITY IN SUPERCONDUCTORS

In attempting to estimate the velocity of second sound in superconductors we are, of course, handicapped by the lack of an adequate detailed theory of their properties. Some progress may be made, however, by starting from some considerations advanced by Gorter and Casimir (1934). Noting that in the absence of an external magnetic field there is no entropy change at the transition point, i.e. $S(T_c)=\gamma T_c$ per unit volume, they assumed that the free energy of a superconductor may be expressed in the form

$$F=-\beta(1-x)-\tfrac{1}{2}\gamma T^2 x^\alpha, \qquad \ldots\ldots(5.1)$$

where x is a parameter varying from 0 at absolute zero to 1 at the transition point. Equilibrium between the 'superconducting electrons' represented by the first term in (5.1) and the 'normal electrons' described by the second requires that $\partial F/\partial x=0$, so that

$$x=(2\beta/\alpha\gamma T^2)^{1/(\alpha-1)}=(T/T_c)^{2/(1-\alpha)} \qquad \ldots\ldots(5.2)$$

since $x=1$ at the transition temperature T_c. The entropy and specific heat obtained from the free energy are, per unit volume,

$$S=\gamma T\left(\frac{T}{T_c}\right)^{2\alpha/(1-\alpha)}, \qquad C=\frac{1+\alpha}{1-\alpha}\gamma T\left(\frac{T}{T_c}\right)^{2\alpha/(1-\alpha)}. \qquad \ldots\ldots(5.3)$$

It is plausible to assume that the parameter x given by (5.2) may be identified with $\rho_n/\rho_{\text{electrons}}$,* in which case the velocity at very low temperatures becomes

$$c_2=T_c\left\{\frac{\gamma}{Nm}\,\frac{1-\alpha}{1+\alpha}\right\}^{1/2}=\pi T_c\left\{\frac{\boldsymbol{k}}{2m'T_0}\,\frac{1-\alpha}{1+\alpha}\right\}^{1/2}=\frac{\pi\boldsymbol{k}T_c}{p_0}\left\{\frac{1-\alpha}{1+\alpha}\right\}^{1/2}. \qquad \ldots\ldots(5.4)$$

In an isolated assembly of electrons c_2 would decrease to zero at the transition point and, according to § 4, remain zero above it (fig. 2). But for electrons in equilibrium with lattice vibrations the relevant total density would be ρ and not Nm, so that c_2 would be very nearly constant up to the transition temperature (fig. 3), since $\rho_n/\rho\ll 1$ in the entire superconducting region.

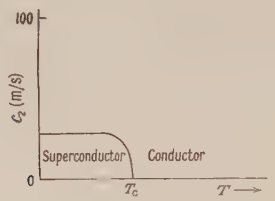

Fig. 2. Isolated electrons.

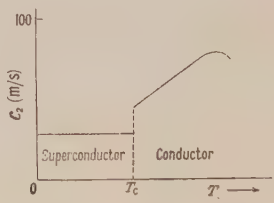

Fig. 3. Electrons within a lattice.

Comparison of (5.4) with (4.5) shows that the velocity in a lattice increases discontinuously by a factor $\{(1+\alpha)/(1-\alpha)\}^{1/2}$ as the temperature is increased past the transition point. Experimentally, the specific heat of superconductors seems to be proportional to T^3, so that $\alpha=\frac{1}{2}$; c_2 should therefore jump by a factor $\sqrt{3}$ at the transition temperature.

In an *impure* superconductor the (residual) ohmic resistance of the 'normal electrons' would probably result in severe attenuation.

* Another possibility would be to identify x with the fractional concentration of normal electrons N_n/N, but then we could not deduce $\rho_n/\rho_{\text{electrons}}$ without further assumptions. For the propagation of second sound within a pure superconductor it is probably necessary that $\rho_n/\rho_{\text{electrons}}\simeq N_n/N$; otherwise the condition of zero net momentum flux at any point (necessary for second sound) would not be compatible with the condition of zero net variation in internal electric current (necessary in a pure superconductor).

§ 6. VELOCITY IN FERROMAGNETS

We calculate first the velocity of second sound to be expected on Stoner's theory of collective electron ferromagnetism (Stoner 1938, 1939), and then that to be expected on Bloch's theory of spin-waves (Bloch 1930). According to Stoner (1939, eqn. 4.22), the entropy and specific heat at very low temperatures are, per unit volume,

$$S=C=\gamma T\frac{(1+\zeta_0)^{1/3}+(1-\zeta_0)^{1/3}}{2} \qquad \ldots\ldots(6.1)$$

where γ is the Sommerfeld coefficient and ζ_0 the relative magnetization near absolute zero. Since Stoner assumes a parabolic form for the kinetic energy spectrum of the electrons, we have $\rho_n=Nm=\rho_{\text{electrons}}$ and second-sound propagation is impossible in the isolated system, just as for an ordinary conductor.

If the electrons are in equilibrium with the lattice vibrations, similar considerations to those of § 4 suggest that

$$c_2=\Xi T(\gamma/Nm)^{1/2}=\Xi\pi T(\boldsymbol{k}/2mT_0)^{1/2}=\Xi\pi\boldsymbol{k}T/p_0, \qquad \ldots\ldots(6.2)$$

where

$$\Xi=\left\{\frac{(1+\zeta_0)^{1/3}+(1-\zeta_0)^{1/3}}{2}\right\}^{1/2}. \qquad \ldots\ldots(6.3)$$

Ξ varies from 1 for $\zeta_0=0$, which occurs if the energy band is very wide, to $2^{-1/3}=0{\cdot}7937$ for $\zeta_0=1$, corresponding to complete saturation.

In calculating the velocity according to the Bloch spin-wave model, we follow the notation of Mott and Jones (1936), except that we change their ρ to x in order to avoid confusion with the density. Taking I to be the exchange integral, we have (Mott and Jones 1936, p. 236)

$$E(p)=Ix^2 \qquad \ldots\ldots(6.4)$$

and the number of states per unit range is, per unit volume,

$$(G/2\pi)^3 4\pi x^2\,dx \qquad \ldots\ldots(6.5)$$

where $G^3=\nu N$, N being the number of atoms per unit volume and ν a numerical factor equal to 1 for a simple cubic lattice, 2 for a body-centred cubic, and 4 for one that is face-centred (Mott and Jones 1936, p. 234). Comparison with the usual expression for the number of states shows that $x=p/G\hbar$, so that $E(p)=Ip^2/G^2\hbar^2$, and the effective mass of the spin-waves must be

$$m'=G^2\hbar^2/2I. \qquad \ldots\ldots(6.6)$$

Since this is independent of momentum, $\rho_n=\mathcal{N}'\boldsymbol{m}'$, where $\mathcal{N}'$ is the number of spin-waves per unit volume given by

$$\mathcal{N}'=4\pi\left(\frac{G}{2\pi}\right)^3\int_0^\infty\frac{x^2\,dx}{\exp(Ix^2/\boldsymbol{k}T)-1}=\frac{G^3}{8\pi^{3/2}}\left(\frac{\boldsymbol{k}T}{I}\right)^{3/2}\zeta\left(\frac{3}{2}\right). \qquad \ldots\ldots(6.7)$$

Thus

$$\rho_n=\frac{G^5\hbar^2}{16\pi^{3/2}I}\left(\frac{\boldsymbol{k}T}{I}\right)^{3/2}\zeta\left(\frac{3}{2}\right). \qquad \ldots\ldots(6.8)$$

The entropy per unit volume is found to be

$$S=\frac{5\boldsymbol{k}G^3}{16\pi^{3/2}}\left(\frac{\boldsymbol{k}T}{I}\right)^{3/2}\zeta\left(\frac{5}{2}\right). \qquad \ldots\ldots(6.9)$$

If, as at low temperatures, $\rho_n\ll\rho$,

$$c_2=\frac{1}{G\hbar}\left\{\frac{10\boldsymbol{k}TI\zeta(\frac{5}{2})}{3\zeta(\frac{3}{2})}\right\}^{1/2}=\frac{1{\cdot}46\times10^{19}(TI)^{1/2}}{G}=1{\cdot}725\times10^{11}(TI)^{1/2}(V/\nu)^{1/3} \qquad \ldots\ldots(6.10)$$

where V is the molecular volume and the units are those of the c.g.s. system. Inserting in (6.10) the values of the exchange integral deduced by Wohlfarth (1949) from Fallot's (1936) experimental results, the velocity for iron is found to be about $790\sqrt{T}$ m/s, and that for nickel about $360\sqrt{T}$ m/s. In these materials, however, these velocities could probably only be observed at such high frequencies that the spin-waves no longer remain in equilibrium with the lattice vibrations, because the spin-wave contributions to S, C and ρ_n are much smaller than those both of the lattice and of electrons in partially filled bands, of which the Bloch theory takes no account. The waves would then have to be excited and detected by magnetic rather than thermal means.

For a satisfactory comparison with experiment at normal frequencies it would be necessary to find a metal or alloy in which there is practically no electronic specific heat linear in temperature, and to perform the experiment at such low temperatures that the Debye lattice contributions are negligible in comparison with those of the spin-waves.

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LETTERS TO THE EDITOR

Relative Pair Production Cross Section at Low Energies

In the course of testing a three crystal scintillation spectrometer for the measurement of γ-ray energies, similar to those reported by Bair and Maienschein (1951) and by Johansson (1952), it was found that the ' pair peaks ' due to γ-rays of energy only slightly in excess of the pair threshold could be resolved. As no experimental data exist for the pair production cross section in the vicinity of the threshold and as the theory using the Born approximation is not strictly applicable, it seemed worth while to measure the relative cross section using the cascade γ-rays from ^{24}Na (2·765 and 1·380 Mev) and from ^{60}Co (1·332 and 1·171 Mev). Since the two gamma quanta from each of these isotopes occur in equal numbers, the ratio of the areas under the two pair peaks gives directly the ratio of the pair production cross sections at the two energies. The internal pair and internal conversion coefficients are not large enough to affect significantly the equality in number of the cascade gamma quanta emitted from sources of these isotopes.

A lead collimator 12 in. long defined the γ-ray beam entering a centre counter consisting of a block of NaI(Tl) 1·4 cm × 1·4 cm × 2·7 cm mounted on a 14 stage photomultiplier

(E.M.I. 6262). This multiplier was operated at 1250 volts for the ^{24}Na runs and at 1400 volts for the ^{60}Co runs to ensure linearity between pulse height and energy release in the crystal. In order to select only those events occurring in the centre crystal in which the γ-rays were converted into pairs (of kinetic energy $h\nu - 2mc^2$) two counters were placed on opposite sides of the centre crystal to detect the two collinear annihilation quanta resulting from the decay of the positron. These counters consisted of cylinders of NaI, 1 inch long by 1 inch in diameter, mounted on 11 stage E.M.I. 5311 photomultipliers operated at 1800 volts. Pulses from these side counters were fed to a coincidence mixer with a resolving time of 0·15 μsec which opened an electronic gate in the output of the centre counter whenever a coincidence occurred; the pulses from the centre counter which were allowed to pass through the gate were amplified and displayed on an 18 channel ' kicksorter '. The kicksorter was set to cover various parts of the spectrum and runs of several hours duration obtained. In all about 70 hours was spent on each isotope and finally all the data so obtained were normalized to an arbitrary peak value for the higher energy pair peak with the results shown in figs. 1 and 2. The total number of counts under the 1·74 MeV ^{24}Na peak was about 82000 and under the 310 kev ^{60}Co peak was about 9500, the background being subtracted in each case.

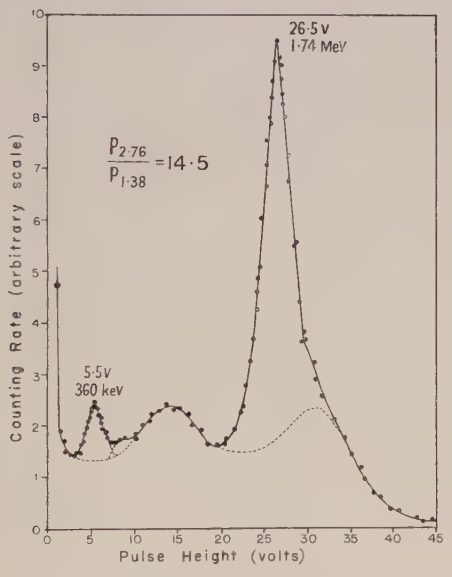

Fig. 1. Three-crystal spectrum of ^{24}Na.

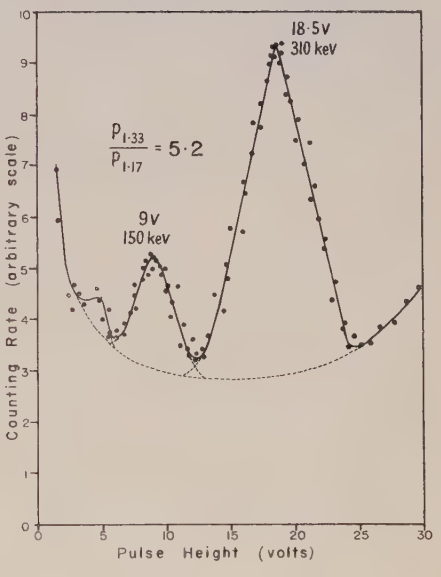

Fig. 2. Three-crystal spectrum of ^{60}Co.

The curves show clearly the ' pair peaks ' superimposed on a background due to accidental triple coincidences, simultaneous Compton scattering of both γ-rays into the two side counters and backscatter from one side counter to the other. For the ^{24}Na runs the side counters were placed 0·3 cm from the centre crystal while for the ^{60}Co the separation was 1·5 cm, so that the background is greater for the ^{24}Na case particularly in the regions of the broad Compton peaks. The ^{60}Co Compton peak occurred near 45 volts and so is not shown in fig. 2. The small bumps at 8 volts for ^{24}Na and at 4 volts for ^{60}Co were introduced by very large pulses, after electronic ' clipping ', passing through the closed gate with much decreased amplitude. The clipping levels were different for the ^{60}Co and ^{24}Na runs.

In order to find the ratio of the areas under the pair peaks it was necessary to subtract the background. For ^{60}Co the shape of the background curve was roughly determined by taking a run with one of the side counters moved out of the direct line of the other two counters so that no ' pair ' coincidences were recorded; a smooth curve was then drawn under the ^{60}Co pair peaks following this shape. For the ^{24}Na no separate determination of the background was made; the upper Compton peak, shown dotted under the 1·74 MeV pair

peak in fig. 1, was estimated from the marked asymmetry at the base of the pair peak, while the background under the lower pair peak was drawn in as shown on the basis of the curve shape on either side of the peak.

After subtracting the background the ratios of the areas under the pair peaks were found graphically to be: $P_{2\cdot76}/P_{1\cdot38}$, experimental $14\cdot5\pm2\cdot5$, theoretical $18\cdot5$; $P_{1\cdot33}/P_{1\cdot17}$, experimental $5\cdot2\pm1\cdot5$, theoretical $6\cdot4$. The experimental errors were estimated from the statistics and from consideration of the background subtraction. The theoretical pair cross section ratios quoted were calculated from the low energy approximation to the Bethe–Heitler formula given by Hough (1948). The discrepancies between the theory and experiment are in the direction to be expected due to the use of the Born approximation. As the cross section has been evaluated only at two energies by Jaeger and Hulme (1935), using a more exact treatment, no quantitative comparison has been made with their theory. However, it would seem that the Jaeger–Hulme theory gives ratios even lower than the above experimental ones.

It is a pleasure to acknowledge the award of an Ontario Research Council Scholarship to one of us (G.M.G.).

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The Forbidden Nature of the Reaction ^{6}Li(γd)^{4}He for γ-Rays of Energies up to 17·6 Mev

In an earlier communication (Titterton 1950) discussing the disintegration of the lithium isotopes by the γ-rays from the 440 kev ^{7}Li(pγ) resonance, it was noted that events corresponding to the reaction ^{6}Li(γd)^{4}He were not observed. An upper limit for the cross section was given as $\sigma_{(17\cdot6+14\cdot8)}\leqslant(0\cdot4\pm0\cdot4)\times10^{-28}$ cm^2. Since that date photographic plate experiments have been carried out with the monochromatic gamma rays from sodium 24, of which only the 2·76 Mev component is above the threshold (at 1·54 Mev). These give

$$\sigma_{2\cdot76}\leqslant(8\pm2)\times10^{-30}\text{ cm}^2. \qquad \ldots\ldots(1)$$

Lately lithium-loaded plates have been exposed to gamma-rays from the ^{19}F(pγ) reaction generated by the 1·2 Mev Cockcroft–Walton accelerator recently installed in this department. A thick CaF_2 target irradiation was made at a bombarding energy of 950 kev. The upper limit for the (γd) cross section for the γ-ray group (6·13, 6·9 and 7·1 Mev) was determined as:

$$\sigma_{6\cdot13}\leqslant(6\pm3)\times10^{-30}\text{ cm}^2. \qquad \ldots\ldots(2)$$

In examining plates loaded with the separated isotope lithium six, which had been exposed to the ^{7}Li(pγ) radiations to study the reaction ^{6}Li(γn)^{5}Li (Titterton and Brinkley 1951), a search has been made for the (γd) reaction. To date no event has been observed and an upper limit for the cross section is found as

$$\sigma_{(17\cdot6+14\cdot8)}\leqslant(5\pm2)\times10^{-30}\text{ cm}^2. \qquad \ldots\ldots(3)$$

In addition to this work with photographic plates two counter experiments have now been performed. Wilson (private communication) finds $\sigma_{2\cdot76}<10^{-31}$ cm^2 and Jensen (private communication) $\sigma_{2\cdot62}<2\times10^{-29}$ cm^2, each of which is compatible with the limit given in (1).

These low values of the upper limits for the cross section are somewhat surprising and the question arises as to the possible reasons for this behaviour.

Many years ago Bethe (1937) pointed out that in systems where the centres of gravity of the neutrons and of the protons coincided the dipole moment would vanish identically whatever the initial and final wave functions. The present reaction ^{6}Li $\rightarrow$ ^{2}D + ^{4}He is expected to be in this category and one might expect dipole transitions to be forbidden. However, the reaction ^{12}C($\gamma\alpha$)^{8}Be, which also satisfies the criterion, has a quite large cross section (about 10^{-28} cm^2 at 17·6 MeV).

Lately evidence has been obtained in this laboratory and elsewhere for the resonance absorption of γ-rays and, accepting the compound nucleus picture, the result could be explained by assuming that, at the γ-ray energies of the experiment, such levels as can be excited are prevented from breaking up into a deuteron and an α-particle by selection rules.

Two levels of ^{6}Li are known, at 2·187 and 3·58 MeV. We have some evidence (unpublished) suggesting that the first of these levels disintegrates into an (α+d). Day and Walker (1952) have observed γ-rays from the 3·58 MeV level and, to explain the preferred γ-emission, conclude that it must have zero spin and even parity, when break-up into charged particles is forbidden. At excitation energies about 4·5 and 5·3 MeV respectively break-up into (p+^{5}He) and (n+^{5}Li) becomes energetically possible and competes with (α+d). These reactions may be strongly preferred. However, against this, Almqvist *et al.* (1951) bombarding a tritium target with 200 kev ^{3}He ions have observed break-up of the compound nucleus into (α+d), as well as (^{5}He+p) and (^{4}He+n+p). This indicates that at an excitation energy of 15·8 MeV the compound nucleus can disintegrate into an α-particle and a deuteron.

It is hoped to carry out experiments at intermediate energies and work with plates exposed to the bremsstrahlung spectrum of a 24 MeV synchrotron is already in progress.

We thank the four observers, Misses Miller, Velioniskyte and Zubans and Mrs. Strautmanis who carried out part of the microscope work and Dr. F. C. Barker for discussion.

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Energy of a Bloch Wall on the Band Picture

In two papers under the above title Herring (1952 a, b) has recently discussed the origin and magnitude of the so-called stiffness constant A determining the characteristics of domain boundaries in ferromagnetic media. The discussion is based on the collective electron approach whereas earlier calculations of A (Kittel 1949) had been made on the basis of the classical Heitler–London–Heisenberg theory.

The calculation of A using the classical theory is based on a simple pictorial representation of a Bloch wall as containing atoms whose magnetically effective electrons have uniformly varying spin orientations across the boundary. The stiffness constant is then directly related to the effective molecular field coefficient ν and if this is expressed in terms of the Curie temperature θ, then (Stoner 1950)

$$A \simeq \tfrac{1}{10} q^{1/3} k\theta / a_1, \quad \ldots\ldots (1)$$

where q is the number of magnetic carriers and a_1^3 the volume per atom, giving $A = 8{\cdot}3 \times 10^{-7}$ erg cm^{-1} for iron. Kittel's (1949) estimate of A for iron, $2{\cdot}0 \times 10^{-6}$, is based on the Bloch spin wave treatment and is more than twice as large as the present estimate. In calculations of A based on the collective electron theory there are many fundamental theoretical difficulties since without much modification this approach is only

designed to treat the properties of a homogeneous ferromagnetic, i.e. a single large domain. One such modification must be the introduction of spin–orbit coupling and the consequent appearance of magneto-crystalline anisotropy (Brooks 1940). This is apparently not taken into account in Herring's work which is largely phenomenological in that no attempt is made to explain *why* there is a gradual change of spin directions of the type envisaged ('spiralling', response to a varying externally applied torque). Formally, a determination of A is possible without appeal to any theoretical model by using the experimental value of the molecular field coefficient ν obtained in principle from magnetocaloric measurements (Stoner 1936). Otherwise, as in deriving (1), ν may be expressed in terms of other ferromagnetic characteristics on the basis of some theoretical model. With the classical theory

$$\nu = N\boldsymbol{k}\theta/I_0^2 \qquad \ldots\ldots (2)$$

where N is the number of magnetic carriers per unit volume and I_0 the saturation magnetization. With the collective electron theory the author has shown (Wohlfarth 1951) that in the special case of rectangular energy bands the molecular field coefficient is modified and becomes ν', where, at 0°K,

$$r_0 = \nu'/\nu = (\theta'/\theta)(1 - \epsilon_0/\boldsymbol{k}\theta'), \qquad \ldots\ldots (3)$$

θ' and ϵ_0 being the usual exchange and bandwidth parameters. Using (3) rather than (2) we find

$$A \simeq \tfrac{1}{10} q^{1/3} \boldsymbol{k}\theta'(1 - \epsilon_0/\boldsymbol{k}\theta')/a_1 \qquad \ldots\ldots (4)$$

at 0° K. The ratio r_0 is calculable from the observed temperature variation of the susceptibility above the Curie point, the author's estimate (Wohlfarth 1951) for nickel being $r_0 \simeq 0{\cdot}24$. Assuming the same value for iron the calculated value of A at 0° K is $2{\cdot}0 \times 10^{-7}$ erg cm^{-1}. The ratio ν'/ν increases almost linearly with temperature, reaching, for nickel, a value $r_1 \simeq 3r_0$ at the Curie point in the absence of a magnetic field. A provisional room temperature estimate of A for iron along these lines is thus about $3{\cdot}0 \times 10^{-7}$ erg cm^{-1}, a value more than five times smaller than Kittel's estimate (1949). In the absence of reliable experimental data (powder patterns, single domain ferromagnetic powders) it seems impossible to choose between any theoretical estimates of A so far made.

Although Herring's expression for A (e.g. the relation (33) in his second paper) contains a factor similar to that in brackets in (4) it differs from (4) by having a factor proportional to ϵ_0 outside the bracket while (4) has $\boldsymbol{k}\theta'$. Using a large value of the 'effective mass' Herring thus obtains an even smaller estimate for A. It is not immediately clear how Herring's relation was obtained and no criticism is intended of his mathematical arguments. It seems a strong point in favour of the present formula (4), however, that it tends towards the classical relation (1) as the bandwidth tends to zero ($\boldsymbol{k}\theta'/\epsilon_0 \to \infty$).

A discussion on the above lines for energy bands of different shapes meets with certain difficulties which may be resolved by work now proceeding on the magnetocaloric effect (Hunt 1952).

Department of Mathematics,
Imperial College, London.
23rd September 1952.

E. P. Wohlfarth.

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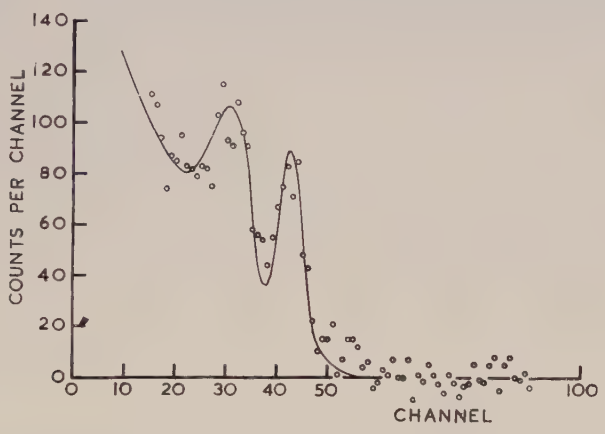
The Radiative Capture of Protons in Fluorine

Devons and Hereward (1948) discovered the radiative capture of protons in fluorine at a proton bombarding energy of 669 kev; Rae, Rutherglen and Smith (1950) and Carver and Wilkinson (1951) showed that the transition probably took place to the first excited state of ^{20}Ne found at 1·5 Mev from ^{19}F(dn)^{20}Ne* (Powell 1942), at 1·4 Mev from ^{20}Ne(pp)^{20}Ne* (Powell, May, Chadwick and Pickavance 1940), at $1{\cdot}63 \pm 0{\cdot}02$ Mev from ^{20}F(β^-)^{20}Ne* (Jelley 1950, Littauer 1950), and at $1{\cdot}63 \pm 0{\cdot}02$ Mev from ^{23}Na(pα)^{20}Ne* (Stelson, Preston and Goodman 1952). We have shown that this is indeed so by measuring the low-energy gamma-ray produced in the cascade.

Two NaI(Tl) crystals in coincidence examined a thick target of CaF_2 bombarded by protons of energy 700 kev; the pulse from one crystal was recorded when the other crystal indicated the emission of a gamma-ray of energy greater than 7·5 Mev. The pulse distribution, corrected for the background of random coincidences, is shown in the figure; also shown is the pulse distribution due to the gamma-rays from ^{20}F(β^-)^{20}Ne* The gamma-ray energy derived from these measurements is $1{\cdot}66 \pm 0{\cdot}02$ Mev.



Pulse distribution in NaI(Tl) from low energy component of gamma-ray cascade following capture of protons of 669 kev in ^{19}F (circles); pulse distribution from gamma-ray following β-decay of ^{20}F (full curve).

The coincidence rate agreed well with that expected on the basis of one low-energy quantum per radiative capture process, assuring us that the cascade is the principal mode and that direct capture to the ground state is relatively rare.

Cavendish Laboratory,
Cambridge.
23rd July 1952, in final form 13th October 1952.

G. A. Jones.
D. H. Wilkinson.

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OBITUARY NOTICES

DONALD KENNETH ASHPOLE

DONALD K. ASHPOLE received his earlier education at Bishop's Stortford College and entered Imperial College in 1942. Two years later he obtained an honours degree in physics and after leaving college spent two years with British Fillers Ltd. He was then appointed assistant physicist at Courtauld's research laboratories at Bocking, but after a short time became senior physicist in charge and held this post up to his untimely death on 3rd March 1952, at the early age of 27 years.

Donald Ashpole's later scientific work dealt with dyeing and moisture relations in textile fibres. He had been invited by the Royal Society to read a paper on this subject and, but for his illness, this would have been delivered by him a few days before his death. In his spare time Ashpole was actively concerned in church work and had sat on the Free Church Federal Council ; he had also produced pageants and plays with much success. To his tutors at college Donald will be remembered for his pleasant and happy temperament and his sincere affection for those who had helped him forward in his scientific career. With his wife and two children we mourn his passing.

R. W. B. STEPHENS.

GRANVILLE HUGH BAILLIE

GRANVILLE HUGH BAILLIE was born in 1873 at Bath. He was educated at a private school (Winton House, Winchester) and entered the City and Guilds College in 1890, winning the Institute's Scholarship. He completed the course in Electrical Engineering in 1893 and was awarded the Diploma of Associate of the City and Guilds of London Institute (A.C.G.I.).

He later attended a post-graduate course at the Hochschule at Zürich. It was there that he met the lady who became his wife.

His first important post was that of Managing Director of the Riviera Electricity Company, Bordighera. He made himself responsible for the installation and maintenance of the complete electrical plant, a position requiring much energy and initiative in a young man of twenty-five.

On his return from Italy he entered into partnership with Mr. James Swinburne, F.R.S. (later Sir James Swinburne, Bart), as a consulting engineer, and they were later joined by Mr. Mervyn O'Gorman. About 1910, this firm having been dissolved, he formed with Mr A. Dobree the firm of Dobree and Baillie as consulting engineers and specializing in patent law.

Baillie's equable temper fitted him eminently for partnership. He and Dobree were both of them exceptional amateur mechanics, Baillie being also a keen exponent of ornamental turning.

Baillie was an excellent draughtsman and drew the majority of the drawings for the patent specifications himself. He often appeared as an expert witness and it was in that capacity that he was the first to use a cinematograph film as evidence in the High Court.

With the outbreak of the 1914–18 war the firm was dissolved and Baillie worked at the Ministry of Munitions on numerous problems connected with the dilution of labour. For this work he was awarded the O.B.E.

After the war Baillie returned to private practice as a consulting engineer. He had been connected with the British Tabulating Machine Company since 1910 and was appointed Joint Managing Director for the period of the war, 1939–45. He acted as Consulting Engineer to the Company until his death.

Early in life his appreciation for fine mechanisms attracted him to a close study of watches, incidental to which he gradually collected an excellent horological library.

In 1929 he produced his first book *Watches, their History, Decorations and Mechanisms*. This was a large and handsome book published by Methuen and Co. in their Connoisseur Library Series. It was followed in the same year, in the same series, by *Watchmakers and Clockmakers of the World*. This contained 25 000 names. In 1947 a second edition of the

latter book was published by the N. A. G. Press containing 35000 names and a great deal more information. The work involved in collecting and sorting out such a number of names was immense.

A few months before his death his *Clocks and Watches, a Horological Bibliography*, was published by the N. A. G. Press. It is perhaps the most authoritative book on the subject, and won for Baillie the Gold Medal of the British Horological Institute.

Baillie rearranged and catalogued the collection of clocks and watches belonging to the Clockmakers Company at the Guildhall, and also the books in the library relating to horology. Whilst engaged on this work he found the manuscript written by John Harrison in 1730 setting out his development of the grid-iron pendulum.

The Company, in connection with the Festival of Britain, issued a small booklet written by Baillie entitled *The Development of Time Measurement*. In this all the line drawings were made by him, and at the exhibition large scale models of the various escapements were made to his design by the apprentices of the British Tabulating Machine Company.

Baillie was a keen motorist and owned his first car about 1900. He was for many years a judge at the official trials of the Royal Automobile Club.

He joined the Physical Society in 1903 and was one of the Founder Fellows of the Institute of Physics in 1920.

As will have been gathered from the above, Baillie was a most versatile person: a first-class engineer, a great worker and an excellent linguist; with it all he was modest and sincere with a great capacity for friendship.

He died at Letchworth on the 19th November 1951, leaving a widow and a married daughter.

R. S. W.

ALLAN HITCHEN FERGUSON

Allan Hitchen Ferguson was, in appearance, not unlike Samuel Johnson, though without the coarseness that seems to have been a prominent feature of the Great Cham of literature. With a height of about 6 ft. 6 in. he weighed over 21 stone, which implies a reasonable proportion between height and girth, for (on a cube law) it corresponds to a weight of $12\frac{1}{2}$ stone for a man of 5 ft. 8 in. It meant, however, that he must of necessity be noticed in any company in which he found himself, and it was not easy for him to walk along the streets without having people turn round to look at him.

He was a man of very wide interests, which included the classical languages, parts of the history of physics and, appropriately enough, Johnsoniana. Like the earlier Doctor, he was much weaker on the Teutonic languages than on those of Ancient Greece and Rome. I well remember the Society receiving an address in Latin, informing us that some University was about to celebrate one of its centenaries and asking us to appoint a delegate to the celebrations. This was agreed, and a member nominated, and the question of an address for him to present to the University from the Society arose. Within a few hours, Ferguson had produced a flowery piece of Latin oratory (which was subsequently vetted by a Professor of Latin, who found nothing to correct) and when someone expressed surprise he merely said: "We haven't all forgotten our Matriculation Latin, you know".

This versatility was evident, too, at the examinations in the University of London, where in later years he examined in Modern History (often in association with F. J. Weaver, late Headmaster of Braintree) as well as in Physics.

Ferguson was born at Entwistle on 11th May 1880 and spent his boyhood there, then at Wyke and Blackpool and finally in Preston. It was here that he started a course of training as a pharmacist's assistant and began to study at the Harris Institute for a London Science Degree. He entered the University College of North Wales, Bangor, as an Exhibitioner in 1902 and duly took the London Honours degree in Physics about 1904.

On graduation he became assistant lecturer and senior demonstrator in Physics at Bangor, where he remained until 1919. During this period he became interested in the mathematics of flight and, with E. H. Harker, wrote a book, *Aerial Locomotion* (with a foreword by Professor G. H. Bryan), which was published in 1911 by the Cambridge University Press in their series of Science Manuals. He took the M.A. (Wales) degree in Mathematics and his London D.Sc. in 1915 for research work in Surface Tension. His period at Bangor of course included the whole of the first World War, during which depletion of the staff left him for two years in sole charge of the Applied Mathematics

division; in addition, for three sessions he had also to be the lecturer in Electrical Engineering. Even with this load of work he found time to be interested in hydro-electric development in Wales, and was an active member of the committee dealing with this matter. He was one of those sent to give evidence to the Water Power Resources Committee of the Board of Trade.

In April 1919 he took a post as Lecturer at Manchester College of Technology, but only stayed there for two years, after which he migrated to London, and in October 1921 became a lecturer in the physics department of Queen Mary College, at that time called East London College. He spent the rest of his career here, first as senior lecturer and, from 1934, as associate professor.

Immediately on coming to London in 1921 Ferguson joined the Physical Society, with which he was to be so intimately connected for more than 20 years. He was elected to the Council in 1925, and acted as Papers Secretary from 1928 until 1938. He was thus in office during many of the main events of the inter-war period, beginning with the amalgamation of the Optical Society with the Physical Society in 1932. He was, if not the prime originator of the idea of the *Progress Reports*, at least one of its main protagonists; he pushed the idea, against the opposition of those who thought it would involve financial loss, wrote to prospective authors, and himself edited the first few volumes. As he had to explain to the authors that long articles, involving a lot of hunting for references, were wanted, and that no payment was proposed for the first issues, his task cannot have been easy. At that time the office staff was small and the Society had no Secretary-Editor; the *Proceedings* were edited by Captain C. W. Hume, who devotedly gave his spare time to this work for many years, but naturally could not take on this additional task. Ferguson had a particular wish to include in each annual volume a report on the teaching of physics, a subject he had much at heart; the writing of it usually fell to him, and his report, though rather incongruous, was scholarly and often provocative. Ferguson also played a great part, with Ezer Griffiths, in organizing the International Physics Conference of 1934, which coincided with a meeting of the Symbols, Units and Nomenclature Commission of the International Union.

During his term as Papers Secretary and President of the Physical Society it became advisable to reduce costs and a change of printer was advocated. Ferguson had been appointed an Editor of the *Philosophical Magazine* in 1937 and had subsequently, in 1938, become a Director of the firm of Taylor and Francis, the owners of the *Philosophical Magazine*. At the suggestion of Wilfred Jevons, Taylor and Francis were asked to tender for the *Proceedings* and it was largely due to Ferguson that very satisfactory rates were obtained.

These inter-war years were, however, not without troubles for Ferguson. He had been having trouble with his eyes for some time, and in January 1933 he underwent an operation for cataract, but he was not long in enjoyment of the better vision which resulted. He noticed a blurring of things in the late summer of 1934, and was able to follow and report a progressive deterioration. He was in fact suffering a retinal detachment, for which he was operated on in October 1934. I remember vividly my visit to him in Moorfields Eye Hospital. He was under strict injunctions not to move his head, so that as one entered the room one saw the massive frame lying on its back, and a voice directed towards the middle of the ceiling asked who it was. He had been very interested in the operation (he was not given a general anaesthetic) and was vastly amused at the idea of electrical soldering applied within the human eye.

After this operation he recovered his vision to a great extent, but he was always a great anxiety to his friends, during his visits to London. We had to pilot him about buildings, tell him where to expect a step, and guide him to his vehicle (it had to be a taxi rather too often for a modest pocket), yet he was always cheerful, ready with an apt quotation, and master of the subjects which he had come to discuss.

I do not know whether it was a deliberate choice, made when he came to London, or whether it was gradually forced on him, but from the age of 40 Ferguson devoted himself to those tasks of organization and committee attendance which are so necessary but so shunned by most scientists. Probably his combination of efficiency and of readiness to turn his hand to tasks that needed to be done made him accept first one post and then another till he had no option but to continue. I have sketched his connection with the Physical Society from 1921 up to the end of the inter-war period (he gave up the Papers Secretaryship in 1938), but he was equally active in other fields of physical organization. He joined the Institute of Physics as a Fellow in 1928, and in the same year began a term

of service on the Board. In accordance with the rules he had to stand down in 1933, but he was back on the Board in 1934 and remained there, as member and then as Vice-president, until 1943. During this time he was active in the cause of unity, or at least of co-ordination, between the Institute and the Society, but in this regard he was before his time. There are still some who are active in this cause, and they will have their success some day, if only because logic demands it, but the time is not yet.

He was a member of the Faraday Society and became a member of its Council (1936–1937) and a Vice-president (1937–1943, with a statutory break of one year); he was prominent as a reviewer of books and referee of papers and, from 1947 until his death, was an Honorary Life Member of the Society.

I do not know just when he became Recorder of Section A of the British Association, but he held the position when I first joined the organizing committee, and I was staggered by the energy he put into the task of making the programmes for the annual meetings coherent. When the Association met at Blackpool in 1936 Ferguson took a great delight in being back on his own home ground, and had made it quite clear that the presidency of Section A at a meeting in that town would give him intense pleasure; so that in 1936 he was prevailed on to hand over the post of recorder to Ezer Griffiths and himself became President of the Section, a well-deserved honour. At that time he became also General Secretary of the whole Association, and retained this duty until 1946.

Just before the war, then, Ferguson had been Papers Secretary of the Physical Society, and had begun to take a major part in the editorship of the *Philosophical Magazine*. This did not end his part in the publications policy of the country at that time. It was in January 1939 that he began work as an Associate editor of *Nature*, and he carried this on into the early years of the war, during which time the proposal for a new journal, *Endeavour*, was mooted. He naturally became a member of the editorial panel of this publication, too.

But it is now time to consider the period of the second World War. Ferguson, on handing the Papers Secretaryship of the Physical Society to me, became President for the two-year period 1938 to 1940, but the outbreak of war presented the Society, like everyone and everything else, with new problems, and it was thought better, at the end of his normal term, to ask him to carry on for a further term, rather than to change Presidents in those early critical days. He therefore filled the office until 1941, the only President in the history of the Society to serve for such a term. The strain, with all these outside activities in addition to his work at College, must have been terrific, and I often feel that we may be in some degree responsible for the afflictions which he suffered soon afterwards. He slipped in boarding a bus and fell heavily, knocking his head; he was very badly bruised but he made light of the matter at the time. However, a type of paralysis gradually made itself manifest, which in 1942 was diagnosed as *myasthenia gravis*. It affected his powers of speech, and at first made it difficult to follow him, but later one became accustomed to it; perhaps too he gained more control. Moreover, he was given tablets which enabled him to gain temporary control for a lecture at college or any special occasion, and when he broadcast in a B.B.C. programme he was quite clear.

During the war Queen Mary College was evacuated to Cambridge, and Ferguson had the joy of lecturing in the Cavendish Laboratory theatre and of mixing with the Cambridge physicists. He retired from his University post in 1945, and was made a fellow of Queen Mary College in 1946. He was still very busy, and made frequent visits to London to attend various committees, remaining a member of the Editorial Board of the *Philosophical Magazine*, and in 1948 he prepared the special Commemoration volume, in which his knowledge of eighteenth century science shone through the whole plan. But his condition gradually became worse. Somewhat later his condition was aggravated by Parkinson's disease, and in 1949 further complication due to inflammation at the base of the oesophagus developed. From that time until his peaceful death on 9th November 1951 he lived on a liquid diet, devotedly tended by his wife Nesta, a former student of his, daughter of Mr. John Thomas, former Vice-Principal of the Naval College, Bangor, whom he had married in 1919. After his paralysis it was in Nesta's handwriting that his characteristic letters used to arrive, for he did not allow such a minor matter as bodily weakness to interfere with his interests and activities, and she certainly would not let him give up if it were merely a matter of writing letters. Ferguson's son, John, had a brilliant career at his University, in which Allan took real pride, and their home was truly a happy Christian one.

Ferguson always preferred to live away from his work. When I first knew him he lived in Ongar, but as London spread he felt that this was not far enough out, and he moved to Bishop's Stortford. At the time of the move he told me how many tons of books had to be packed and crated; I think the number was five, but cannot be quite sure. At any rate, shelves were put in all the rooms for their reception. He used his journeys for reading, and in the Ongar days he had a spell of interest in detective stories, of which he reckoned to read one a day in the train. But this was not all his reading. He had, as has been said already, a deep interest in eighteenth century literary life, and all this was not to the exclusion of his scientific reading. He was an expert in classical physics, particularly surface tension, and knew a surprising amount about modern developments.

This was fittingly recognized by the Editors of the *New Chambers' Encyclopaedia*, who asked him to organize the articles on physical subjects. It reminds me that he must be one of the few people to review the *Encyclopaedia Britannica*; when the new, post-war edition came out he reviewed it for our *Proceedings*, not confining himself to the scientific articles, but commenting on its treatment of other subjects, and pointing out that it had no reference to the Papworth settlement for the tuberculous.

In politics Allan Ferguson was a Liberal, of the Manchester school, but he had a good deal of sympathy with the Labour movement, and an appreciation that there was much that is valuable in the doctrines of the traditional enemies of the Liberals, the Conservative party. He believed very strongly in the League of Nations, and was a leading light of the local branch of that organization. The failure of the League when its testing time came was a source of grievous disappointment to him.

Thus in his last six years, after his retirement, he had, despite his bodily discomforts, some consolations: a devoted wife, helping him to keep up his interests in the world of physics; a clever, loving son of temperament not unlike his own; he had his books, though he could not always read very much, and he had his correspondence with old friends. Letters, and visits from them, were a source of great consolation to him, and he was always more energetic after a group of scientific colleagues had been to see him. The end was a peaceful one, and the service at Bishop's Stortford preceding his cremation was attended by many of those who had loved and honoured him.

J. H. AWBERY.*

* I should like to express my appreciation of the help which Dr. W. Jevons has given me in making this a less unworthy account; he has reminded me of some incidents which had slipped my mind and told me of others unknown to me which I am glad to see placed on record.

MEYER FINN

Mr. M. Finn died on 8th February 1952 in his eightieth year; he had been a Fellow of the Physical Society for some fifty years. He was born of Jewish parents in Bialastock on the Russian side of the Polish border and managed to obtain a place in the Russian Gymnasium although only one per cent of the available places were then allocated to Jews. He came to England when he was seventeen and entered Armstrong College in the University of Durham. In three days he learnt enough English to attend lectures and finally became a distinguished mathematical scholar of the University, holding a Junior Fellowship for a time. Mr. Finn went to Southend in 1897 to take up work at a small school for boys and girls which eventually became Southend High School for Boys. It was during a Practical Physics Examination carried out for the Oxford and Cambridge Joint Board many years ago that I came into contact with him and was very much impressed by the fact that he had designed nearly all his electricity and magnetism experiments to work from the direct current mains. These experiments were described in his book *Electricity and Magnetism*, published by Bell, now, unfortunately, out of print.

As well as being Senior Master in the day school he was in charge of evening classes at the Southend Evening Technical Institute, where he trained many students for the London B.Sc. Degree, and organized comprehensive curricula in commercial subjects, cookery, dress-making, etc. He retired at the age of sixty-five, but during the war returned to teach Physics to sixth form boys of St. Paul's School and the Dagenham Secondary School.

Immediately after his arrival in England Finn became a naturalized British subject and was equally proud of his Jewish birth and his British nationality; he married an Englishwoman and they had eleven children, of whom seven are in this country, three in the United States and one in Australia. His wife died in December 1946 soon after their golden wedding, but he lived to see his great grandson.

L. F. B.

Allan Ferguson.

JULIUS HARTMANN

By the death of Julius Hartmann on 6th November 1951 Danish science and technical research has suffered a great loss, and the Technical University of Denmark (T.U.D.) has been deprived of one of its most active teachers.

Hartmann's whole career as a scientific worker was intimately connected with and devoted to the development of his Laboratory of Technical Physics at the T.U.D. Already as a young student he began scientific work as Professor K. Prytz's assistant in the old Physical Laboratory. In 1914 he became a lecturer (Docent) of Physics, and in 1929 he was made a professor of Technical Physics, which latter discipline he had introduced as a new educational subject of the T.U.D. Ten years later a well-equipped laboratory—thanks to Hartmann's indefatigable efforts—was established in the new buildings then erected for the T.U.D., and here he spent the last years of his very active lifetime.

Hartmann's scientific work was mainly centred on problems raised by and connected with a series of inventions and discoveries most of which he had made during his younger years. Here may be mentioned his jet-wave commutator and rectifier for alternating current, which is described in a paper *Nye Ensrettere og periodiske Afbrydere* (1918), for which he obtained, as the first in Denmark, the Dr.techn. degree at the T.U.D.

Hartmann tried, with more scientific and technical than economic success, to develop the jet-wave rectifier into an industrial apparatus. In two big monographs, *The Jet-Wave Rectifier* (1929) and *The Large Capacity Jet-Wave Commutator*, I–II (in collaboration with L. Løgstrup Jensen, 1944 and 1946) and a number of smaller papers, Hartmann has given the results obtained by this work. Many new and important observations and inventions relating to fluid flow were made, such as the ' Plate-Jet ' and the electrodynamic mercury-pump.

When trying to measure the pressure-distribution in an air-jet of supersonic velocity by means of a Pitot-tube Hartmann observed pressure fluctuations in the Pitot-system. A closer investigation of this phenomenon led to Hartmann's construction of a generator for very intense sound-waves, described in *The Acoustic Air-Jet Generator* (1939) and now generally known as the ' Hartmann Acoustic Generator ' and mentioned in all modern textbooks of acoustics. In many laboratories it has been used as a most convenient and practical source for high-intensity sound waves.

Hartmann has described many interesting and important phenomena produced by use of his generator, such as the association and precipitation of particles in a colloidal suspension and, under certain other conditions, the opposite effect, production of emulsions. Certain biological effects, as, for instance, disinfection of liquids containing microbes, have also been observed.

For the detailed investigation of the structure of a pulsating air-jet Hartmann, in collaboration with F. Larris, developed a special Schlierenmethod which gave very nice photographic results.

As a teacher of physics at the T.U.D., where the Danish engineers (Civilingeniører) are educated, Hartmann's endeavours were directed mainly on the experimental work and especially on the techniques of measurement and evaluation of observations. He wrote several textbooks on these subjects; the last of these came from printing just a few days before his death.

Personally Hartmann was a kind and modest man, who preferred work to most other forms of human enjoyment of life. His many friends and admirers in his own country and abroad deeply regret his death.

R. E. H. RASMUSSEN.

CHARLES HERBERT LEES

By the death of Professor C. H. Lees the Physical Society has lost its oldest past president and the world of physics will mourn the departure of one of the 'old guard ' of the last generation. He was 88 years of age and died on September 25th 1952. Lees was born in Glodwick, Oldham, in 1864, the second of three sons of Jane and John Lees. John Lees was one of the gold seekers who went to Australia in the '50s. He and his partner found the " Leg of Mutton " nugget at Ballarat. With the proceeds John Lees founded the Ballarat building in Oldham and C. H. Lees as a young man was always interested in the upkeep and repair of the property.

Lees left school at the age of 14 and went as a clerk to the Gas Company's office in Oldham. He attended night school and gained a scholarship to the Owens College, Manchester. When he took his degree he was a Derby Prizeman and elected Bishop Berkeley Fellow in 1888 : on an average about three of these were awarded each year from 1881 to 1895, after which they were discontinued. They were open to all Faculties and Lees was one of only five physicists elected to fellowships during the 16 years. He studied at the University of Strasbourg for about two years and then spent a few months teaching at a school. His Strasbourg professors were Kundt in physics and Christoffel and Rye in Mathematics. In 1891 he returned to Manchester and was appointed senior assistant lecturer and demonstrator in Physics in the Owens College and became lecturer and assistant director of the Physical Laboratories of the University of Manchester in 1900. He worked under Schuster. Until his marriage in 1902 Lees spent much of his holiday time walking and geologizing in Lancashire and Yorkshire, often accompanied by his younger brother. He left Manchester in 1906 to become professor at Queen Mary College, then East London College. Just before he left Manchester he went to South Africa as secretary of Section A (Physics and Mathematics) of the British Association. He served Queen Mary College until 1930 retiring with the title of Emeritus Professor of Physics in the University of London. He was Vice-Principal of the College from 1918 to 1930 and a representative of the Academic Board on the Governing Body from 1916 to 1930. It is certain that the Principal, J. L. S. Hatton, relied a great deal on Lees' judgment in college administration and academic matters. In his earlier years at the college he had one lecturer to share the work and there were evening as well as day classes. By 1929-30, his last session, his staff had increased to four lecturers and the evening classes had been abolished. Throughout the time Lees took a large share in the lecturing to Intermediate, General B.Sc. and Special Honours students.

It was in 1907 that the College first obtained recognition as a school of the University of London for an initial period of three years. This was extended for a further five-year period in 1910 and finally without limitation in 1915. Lees' part in developing the Physics Department and his general influence on college affairs must have contributed much to this.

He was one of the first three to be elected Fellows of Queen Mary College when it received its new charter in 1934. For many years Lees served on the Kent Education Committee and the Delegacy for Managing Goldsmith's College.

Lees' major contribution to physics was in the field of thermal measurement. During his stay at Strasbourg he carried out some experiments on thermal conductivity by the 'periodic' method first used by Ångström. He found that the variation of temperature along the bar could not be represented by the expression deduced by Ångström from Fourier's equation for the flow of heat along a bar. On his return to Manchester he made a theoretical study of the problem and published a paper "On the Law of Cooling, and its bearing on the theory of the Motion of Heat in Bars". This was published by the Manchester Literary and Philosophical Society 1889–90.

Three papers were published in the *Philosophical Transactions of the Royal Society* in 1892, 1905 and 1908. The last of these, on "The Effect of Low Temperatures on the Thermal and Electrical Conductivity of Certain Approximately Pure Metals and Alloys", constituted the Bakerian Lecture for 1908. (In this latter work he used resistance thermometry and acquired first-hand knowledge of the use of this method of measuring temperature by working with E. H. Griffiths.) In these papers Lees displayed his mastery of mathematical technique when dealing with the calculation of correction terms. Lees' method for the measurement of the conductivity of materials in the form of discs is now usually found in modern textbooks.

His interest, however, was very wide as will be seen from the titles of some of his other papers.* In 1901 he published a paper in the *Proceedings of the Physical Society* "On the Viscosities of Mixtures of Liquids and of Solutions". In 1929 he contributed to the Society a paper on "The Free Periods of a Composite Elastic Column or Composite Stretched Wire". This arose out of some investigations carried out at the Safety in Mines Research Stations at Sheffield, Lees serving on the Safety in Mines Research Board. His presidential address to the Physical Society was "On the Mechanical Equilibrium of a Sphere of Gravitative Fluid". He also contributed papers on a.c. bridges and on fluid flow through pipes. Lees took a delight in the geometrical or graphical representation of results.

* A complete list of his published papers is available at the offices of the Physical Society.

The writer's first contact with Lees came early in the formation of the Engineering Committee of the Food Investigation Board now the Food Investigation Organisation. Sir William Hardy, with his sure instinct for selecting the right man persuaded Lees to serve on this Committee, which had Sir Alfred Ewing as its chairman and also on the Insulation Sub-Committee.

The first report they produced was on the literature of refrigeration, this was followed by a critical study of insulated rail vans. They drew attention to the disadvantages from the thermal insulation aspect of the considerable number of metal bolts and metal components penetrating the insulation. The point of their criticism will be appreciated when it is remembered that the conductivity of iron is about 1000 times that of cork. For a period of over twenty years the Engineering Committee continued to function. It supervised the work of Professor Frewen Jenkins at Oxford on the properties of refrigerants; it encouraged work on insulating materials, humidity and various other questions at the National Physical Laboratory ; under its auspices the overseas transport of fruit and meat was investigated. One of its efforts in full scale research was the purchase for the Board of a Thames barge, insulating the holds with various materials and equipping it with a refrigerating plant which could be operated from the engine when this was not required for propelling the barge. As an experiment the venture was a success and a profit was shown, but the conservative attitude of those engaged in this trade retarded its adoption.

Lees was president of the Physical Society from 1918 to 1920, and was also a vice-president of the Institute of Physics. His hobby was handicrafts and to within a few days of his death he occupied his time doing carpentry. He was a man of great patience and was not to be deviated from the course he had set himself by difficulties when carrying out a piece of research. When dealing with research students, if unable to give immediately the complete answer to a question he never failed to throw some light on the problem or make valuable suggestions for the solution. He endeared himself to colleagues and students by his great charm and sincerity and we mourn the loss of a staunch friend. He is survived by his widow and five children, all of whom are associated with scientific work. His eldest daughter is the wife of Professor G. L. Brown, C.B.E., F.R.S. EZER GRIFFITHS.

WILLIAM LETHERSICH

WILLIAM LETHERSICH, who died on 2nd March 1952 at the age of 46, gave an example of achievement in the face of physical handicap. Educated at the Tottenham County School he entered Queen Mary College (London) in 1925, securing in 1928 Second Class Honours in Physics, in spite of his indifferent health. After spending a year on research in the Physics department he transferred in 1929 to the Electrical Engineering department under Professor MacGregor-Morris, where he introduced the double balanced calorimeter method of measuring dielectric losses at high frequencies based on the principle of the rate of temperature rise. The method is still in use. In 1930 he joined the staff of the Electrical Research Association, where he remained until his death. At first he worked with Dr. H. G. Taylor (now Director of the Welding Research Association) on the heating of lamp fittings and developed the Schlieren method of displaying and analysing hot air currents for this purpose, inventing certain ameliorative measures. His methods have been widely developed and his improvements are incorporated in modern design. Regarding the risk to wooden poles for overhead lines with abnormally wet heart wood, in the event of insulator failure, he elucidated the mechanism and showed the risk to be slight and capable of avoidance by a very simple expedient. He then began his main scientific work, which was on the rheological properties of insulating materials. He gave a definition of plastic and brittle failure of bitumen, together with a quantitative relation between tensile strength and rate of loading. His greatest achievement, on which he was working when his last illness began, was probably his application of the theory of linear phenomena to rheological properties. He showed that the behaviour of a large number of materials under shear stress could be quantitatively expressed in terms of a simple spectrum of relaxation times, over a range of time intervals from the order of 10^{-5} to 10^{4} sec, 9 decades. In this work he produced several ingenious experimental devices, described in his publications and exhibited at the Physical Society's Exhibition and elsewhere. Full recognition was rather slow in coming to Lethersich on account of a rather obscure literary style, but in the last few years his reputation for theoretical acumen and elegant experimentation had grown rapidly. He was a Fellow of the

Institute of Physics and an Associate Member of the Institution of Electrical Engineers, as well as a well-known member of the Rheologists' Club. He published several papers in the *Proceedings of the Physical Society*, *Journal of Scientific Instruments*, *Proceedings of the Institution of Electrical Engineers* and *British Journal of Applied Physics*.

Lethersich was not just a scientist, he was an accomplished musician and a man of religious principles, esteemed by a wide circle of friends. He was a bachelor who devoted himself to the service of his community. He gained first prize for the organ at the Stratford Music Festival and acted as Organist and Choirmaster at Queen's Hall Methodist Church, Hayes, for 17 years. Frequently he took duty for his former teacher, Mr. Edgar Humphreys, at Bow Church. The heart trouble which led to his death was a serious handicap throughout his life. In spite of it he made permanently valuable contributions to all the scientific fields of his professional endeavour, in the arts he was admired, and as a friend and teacher he was loved.

S. WHITEHEAD.

FREDERICK HENRY NEWMAN

FREDERICK HENRY NEWMAN was born on 8th June 1891, and was educated at the Bath Technical College (Secondary School) from 1904 to 1911, and at the Royal College of Science, South Kensington, from 1911 to 1914 as the holder of a Royal Scholarship. In 1913 he graduated with First Class Honours in Physics and obtained the Associateship of the Royal College of Science. He came under the direct influence of Lord Rayleigh and Professor A. Fowler as a research student during the session 1913 to 1914. He then proceeded to University College, Aberystwyth, as Lecturer in Physics, and in 1915 carried out some research on x-rays, in the course of which his left hand was very severely burnt, which resulted in a wound that never healed. This was a source of much discomfort fo the rest of his life. He obtained a commission in the Forces and served in a technical capacity from 1915 to February 1919, when he retired as Captain.

In March 1919 he was appointed to the only lectureship in Physics in the former Royal Albert Memorial College in Exeter. He then had but a few students, and of the three whom he prepared for the London B.Sc. (Special) degree, two obtained First Class Honours in Physics. This was an augury of the excellent work he was to do at Exeter. He remained a lecturer until 1922 when the College was placed on the Treasury List by the University Grants Committee as the University College of the South West, and in it he became the occupant of the newly-created Chair of Physics. During his tenure his Department grew in every way, so much so that its present teaching staff of six is fully occupied. He was largely responsible for the design and equipment of the Washington Singer Laboratories, opened in 1931, of which he became Director.

Professor Newman held the degrees of D.Sc. and Ph.D. of the University of London (he was the first external student to receive the London Ph.D. in Physics), the diploma of membership of the Royal College of Science and Technology (D.I.C.), was a Fellow of the Institute of Physics, of the Physical Society, and of the Institute of Radiology. He was a member of la Société Francaise de Physique and A.M.I.E.E.

His research work, which was begun at the Royal College of Science, first dealt with the absorption of gases by various metals that had been deposited on the cathodes of electric discharge tubes. This work he resumed at Exeter after World War I, and it subsequently developed into extensive spectroscopic investigations of vapours of the alkali metals and mercury and of the gases, hydrogen and argon, contained at low pressures in discharge tubes at various temperatures. His interest in physical research never waned, although his output of published work in recent years inevitably became less owing to the encroachment on his time of administrative work. Newman also directed the researches of numerous students for whom he chose themes other than Spectroscopy, and the results of their work have been published under their names, so that his influence in the discovery of new knowledge extends well beyond his own particular field of research.

Dr. Newman was the author of three monographs on Physics, viz. *The Production and Measurement of Low Pressures* 1925; *Electrolytic Conduction* 1931; and *Recent Advances in Physics* 1931. In collaboration with one of his former students, V. H. L. Searle, he wrote a standard text-book on *General Properties of Matter* which passed into its fourth edition in 1948 and has since been reprinted several times.

On the outbreak of World War II he was both Chairman and Secretary of the Joint Recruiting Board of the South Western Area, and it was not until 1947 that he relinquished the Secretaryship. On the cessation of hostilities he undertook yet more voluntary work and became Chairman of the Exeter Disablement Advisory Committee of the Ministry of Labour. For many years he was a Scientific Consultant to the Exeter City Police, and in this capacity gave many lectures at police conferences on Scientific Aids in the Detection of Crime and acted as an expert witness in many cases. During the recent war he acted as adviser to one of the Services. He also organized intensive courses for Radio-bursars of six months' duration, of training in Radio Communication over a period of three years, and towards the end of the war he directed two one-year courses for Cadets of the Royal Corps of Signals. Recognition of this public service came in the form of the C.B.E. in the New Year's Honours List of 1949. Since then he was appointed as one of the three West Regional Scientific Advisers to the newly-created Civil Defence Service.

Following the resignation of Dr. John Murray from the Principalship, Professor Newman undertook the dual task of Acting Principal of University College, Exeter, with nearly a thousand students, and that of running his own Department. At the present stage in the development of the College the work entailed by the combined posts was colossal, but Newman managed to cope with the task with an efficiency which characterized everything he undertook.

His powers as a teacher and research worker were considerable. He was extremely popular with his students, both present and past, who frequently sought, and were benefited by, his advice. By nature he was quiet and reserved. Despite the very active life which he led he was never physically robust, and owing to a life-long disability, coupled with the x-ray injured hand, he was frequently in great pain, although he took especial care not to allow the fact to become apparent to others.

His death on 15th April 1952 was comparatively sudden, and in his passing the University College of the South West and the City of Exeter have suffered a great and severe loss. He is survived by his widow, a daughter and a son.

H. T. S. BRITTON.

MALCOLM MACLEOD NICOLSON

Malcolm Macleod Nicolson, Lecturer in Physics at Leeds University, was killed in an accident at a railway station near his home on 21st December 1951. He had been to meet some friends who had been delayed by the thick fog, and whilst leaving the station by a private crossing was struck by a passing train and killed instantly.

He was born in Skye on 6th July 1919, but his family later moved to Lancashire and he entered Manchester University in 1936, graduating with First Class Honours in Physics in 1939. He worked as a research student under Professor Hartree and took an M.Sc. in the following year.

After a short period of service in the Royal Navy, followed by work on radar development, he was appointed in 1941 as lecturer in the Gunnery and Mathematics department of the Royal Military College of Science. In 1945 he went to Cambridge, where he worked under Dr. Orowan on surface phenomena in ionic crystals, work for which he was awarded a Ph.D. in 1948; whilst at the Cavendish Laboratory he also carried out theoretical work on thermionic valves under the direction of Professor Hartree.

Nicolson came to Leeds in October 1948, and in the brief three years he was there he made himself an integral part of the life and work of the Physics Department. Although primarily a theoretical physicist, he took a very active interest in the experimental work in the teaching laboratories, and his lectures were marked by the thoroughness with which he covered his subjects. At the time of his death he had nearly finished writing a book on mathematics for physicists. Arrangements are being made for the completion and publication of this book.

Occupied though he was in teaching and writing, he was generous to a fault in the time he would devote to problems brought to him by colleagues and research students. The loss of a young scientist of great promise, able and enthusiastic, with high standards and sound critical judgement, is a heavy one, and his many friends will sadly miss a delightful companion, lively and forceful in discussion, humorous and kindly, and utterly unselfish.

In 1942 he married Miss Phyllis Lockett, whom he leaves with two young sons.

R. S. T.

REVIEWS OF BOOKS

A Hundred Years of Physics, by W. Wilson. Pp. 319. (London : Duckworth, 1950.) 21*s*.

It is an unforgivable remissness to delay the review of scientific works, particularly one of Physics, for their value is usually strictly temporary and most are out of date before they are printed. My excuse in this case is that here we have a book which will be of permanent value to the students of physics and, as it deals with a hundred years of the subject, will not suffer from waiting two or three for a review.

Most books on the history of science tend to be a catalogue of dates and discoveries interspersed with biographical details more or less relevant to them. They are necessarily unintelligible to anyone not knowing the subject thoroughly. Professor Wilson has not followed this tradition. Indeed he has written on the last hundred years of physics hardly bringing in history at all. It is idle to look in this book for a critical account of the development of physics from 1850 to 1950 or to learn anything about the men who were responsible for its chief advances. Nor will anything be found of the applications of physics or its contribution to the present atomic age, nor even to the influences of industry or war on the advance of physics. For all such information we must look elsewhere in the wide though scattered literature on all these subjects.

What we will find in Professor Wilson's book is a description, on a level suitable to a first year university student, of the actual advances in physics themselves, arranged in a logical rather than historical order. Thus it provides a new, attractive and, at the same time, sound introduction to physics, not as a set of dogmas of accepted knowledge, but as an interlinked series of successive approximations plainly unfinished. He follows out three main themes: thermodynamics, including the derivation of the first and second laws ; the electromagnetic theory of light, and modern physics of the quantum theory and relativity. Anyone following his chapters, which are copiously supplied with references to original papers, will be able to learn physics intelligibly set out as it was built up.

There is, in addition, a most valuable bibliography. Of particular value are the simple descriptions, experimental and mathematical, of the main advances. They can be used as they stand as a basis for teaching.

Of course the picture is not complete ; for brevity's sake branches and links, often important ones, are altogether omitted. There is a notable lack of description of the purely technical advances which made the scientific discoveries possible. The index does not contain the words vacuum or valve. But for what it sets out to do *A Hundred Years of Physics* is pre-eminently successful. It should find its place in every teaching laboratory of physics and the wise lecturer will want to look at it again each time he prepares his courses.

J. D. BERNAL.

Science in Progress, Sixth Series, edited by G. A. Baitsell. Pp. xv+322. (New Haven : Yale University Press ; Oxford : University Press, 1949.) 40*s*.

The progress of science moves with an accelerating pace, but the communication of the results is as slow as, if not slower than, ever. Most of the essays which compose this book were delivered in 1947. The book was published in 1949, reached this country in 1950 and is being reviewed, owing to the inexcusable dilatoriness of the reviewer, in 1952. It would have been useless, even without that, to discuss these essays as the latest fruit of scientific advance. That they are worth reading now, and most of them are, is due to more intrinsic and time enduring factors. All are well and clearly written and, though subsequent discovery has added to the picture in many places, as records of achievement they still hold their own.

The authors of the physical articles, Smyth, Wheeler, Lawrence and Seaborg, are all renowned masters of their field in American science, as are those in the chemical and biological fields, Pauling, Zechmeister, Stanley, Dubos, Beadle, Sturtevant and Kellog. Most are the directors of vast new laboratories with the super apparatus which has been a magnet to scientists all over the world. What they have to say is the real matter of the

history of science. Indeed, Smyth's article *From X-Rays to Nuclear Fission* is ostensibly history and interesting for the critical comparison of the science of 1942 with that of 1896.

" The chain reaction, the next and last discovery to be metnioned, was made under circumstances superficially very different—in secret, in a war laboratory, heavily financed by the United States Government. Even now, a complete quantitative description of the chain reaction has not been published. The beauty of Roentgen's paper or of those on fission was that anyone who read them could repeat the experiment. This is perhaps the most fundamental principle of science ; its methods of operation are so objective and its means of communication so precise that its results are reproducible. Each man can build on the other's work so that knowledge is advanced by a vast cooperative movement.

But we cannot work in this way when atomic bombs may be the reproducible result in a world that is politically unable to control them. Hence, we have to violate our fundamental principles and keep silent about numbers and dimensions." (Page 20.)

" Up to the time that Fermi and his group first made the nuclear reaction go, the uranium project was essentially scientific in aims and methods. December 2, 1942, represents the end of the epoch of atomic physics that began in 1895. Whether the effort of scientists in this period will ultimately benefit mankind or destroy it remains to be seen. The power of the scientific method has been amply demonstrated." (Page 21.)

This is certainly a fact, but its consequences inside and outside science have yet to be fully appreciated. Lawrence on *High Energy Physics* will long be worth reading. Though the achievement of a mere 200Mev in the Berkeley Synchro-Cyclotron seems now small beer in the Bevatron area, the story of it is like a new voyage of discovery, though it must have cost several hundred times what it did to discover America in the first place. In the same way, Seaborg's story of *Eight New Synthetic Elements* will stand, however many more have still to be described or concealed.

So much for the part of *Science in Progress* devoted to physics. In the remaining field the physicist can learn of the great unification of chemistry and biology in the study of enzymes, viruses and microbiology generally. Here the picture is naturally far more complex, but the methods are increasingly the quantitative approach that derives from physics, and advances are almost as rapid. They have, however, a way of being less secure partly due to an over-simplified carry-over of ideas from a different field, and it is probable that the biological section of *Progress* already needs more revision than the physical.

J. D. BERNAL.

Miscellaneous Physical and Chemical Techniques of the Los Alamos Project, edited by ALVIN C. GRAVES and DAROL K. FROMAN. Pp. ix+323. (London: McGraw-Hill, 1952.) 34*s*.

This is the third volume of Division V of the National Nuclear Energy Series, dealing with some of the techniques which had to be developed at the Los Alamos Laboratory to solve scientific problems associated with the development of the atomic bomb. Its value lies in the precise instructions which are given to achieve a certain aim, in the great deal of thought and explanation why a certain operation is done or suggested, and in the assurance that the methods proposed have been amply tested and used quite often by non-scientifically trained personnel. It is because of the nature of the matter that many of the devices were meant for a special problem, but most of them will prove useful to the research worker. It is therefore a book which, like its predecessors, *Electronics*, by W. C. Elmore and M. Sands, and *Ionization Chambers and Counters*, by B. Rossi and H. Staub, will find its way into many research laboratories. The following are the main topics dealt with, Chapter 1, Preparation of Foils, by R. W. Dodson, A. C. Graves, L. Helmholz, D. L. Hufford. R. M. Potter and J. G. Povelites, describes thin deposits of B, U, Th, Np, Pu, Am, LiF, hydrogenous layers on various carriers, their assaying and the handling of micro-quantities of rare substances. Anyone who has spent time making such uniform deposits knows of the difficulties and is grateful for the information. Chapter 2, Neutron Sources, by A. C. Graves, R. L. Walker, R. F. Taschek, A. O. Hanson, J. H. Williams and H. M. Agnew, discusses sources based on alpha bombardment of beryllium, photoneutron sources and the various reactions involving accelerated charged projectiles, with their characteristics, neutron yield and details of their construction. The calibration of neutron sources, flux measurements, and methods of determining the yield from very weak neutron emitters are

extensively described. A specially valuable chapter deals with BF_3 proportional counters; the preparation of pure BF_3 gas concludes this section. Chapter 3 gives a very clear and instructive exposé of the betatron by its first successful designer, D. Kerst. The Modulation of Beams, by B. D. McDaniel, Leo S. Lavatelli and Elizabeth Graves, from ion accelerators such as high voltage equipment or cyclotrons, mostly used in time-of-flight research for neutron studies, has become one of the most powerful methods in the energy region between thermal and a few kilovolts. A comprehensive account of the authors' experiences in this field is given in Chapter 4 and ought to be studied by all engaged in such work. The following chapter, 5, by Thoma Snyder and David Lipkin, deals with high vacuum engineering, giving workshop drawings of diffusion pumps and performance data, satisfactory high vacuum valves, and devices to allow manipulation in high vacuum spaces. Some, but not all of it, is now common knowledge. A last chapter, 6, by J. E. Mack, on Optical Methods and Instruments, falls outside the interest of the nuclear physicist, but is valuable for those who are engaged in studying rapidly changing luminous processes, such as the investigation of the propagation of explosives. It deals mostly with high-speed photographic cameras.

E. BRETSCHER.

French–English Science Dictionary, by LOUIS DE VRIES. Pp. xii + 596. (London : McGraw-Hill, 1952.) 2nd edn. 55*s*. 6*d*.

Professor de Vries has already done great services for workers in science, and one has come to expect his dictionaries to be competently prepared. It is thus no surprise to find this the case with the second, enlarged, edition of his *French–English Science Dictionary*. The ' enlargement ' takes the form of a supplement of terms used in aeronautics, electronics and the trio radar, radio and television.

The merest beginner need have no fears about using this work. The parts of many irregular verbs are given separate entries (' va ' and ' vais ' under ' V ', for example !), the gender of each noun is indicated, and even plurals such as ' chevaux ' are given.

The original volume contained 43 000 words. The supplement brings another 50 pages, and this is followed by 12 pages of abbreviations together with the full words and their English equivalents.

The words are chosen from agricultural and biological subjects as well as from the physical sciences. Nevertheless physics seems to be well represented, especially now that the supplement has been added. The book is well set out and pleasant to use.

H. H. HOPKINS.

Annual Review of Nuclear Science, Vol. I, 1952. Pp. ix + 645. (Stanford, California : Annual Reviews Inc., 1952.) $6.35.

This volume, which deals almost entirely with work published in 1950, is the result of a decision by the Committee on Nuclear Science of the National Research Council of the U.S.A. to sponsor the preparation of a series of Annual Reviews of Nuclear Science. It has been edited by the Sub-committee on Publications, which includes many well-known names. The contents of the second volume have been announced, and we may confidently expect that the *Annual Review* will be a permanent addition to scientific literature. It is natural to begin by making a comparison with the annual *Reports on Progress in Physics*, published by our own Society, and with the *Reviews of Modern Physics*, produced by the American Physical Society, and here one is at once struck by the difference in the field covered. Judging by the contents of the first volume, the *Annual Review* is expected to deal not only with all those subjects which contribute to an understanding of Nuclear Physics and Atomic Energy research, but also with all those subjects whose advance is accelerated by the availability of radioactive and stable isotopes. We find, accordingly, that only about one-half of the book is devoted to physics, and that there are important contributions from chemists, metallurgists, geologists, biologists and medical scientists.

The articles on physics include contributions on The Nuclear Reactions of Cosmic Rays, Meson Physics, Recent Developments in the Theory of Nuclear Structure, The Energy Levels of Light Nuclei, Nuclear Moments, Isotopic Abundances, The Detection of Nuclear Particles, as well as a group of five articles on High Energy Accelerators. There are two other contributions on physical subjects, which, because they appear in this book, deal only

with those aspects of their subjects which are concerned with nuclear science. The article on Atomic and Molecular Spectroscopy deals only with those researches which have some relation to the mass or structure of the nucleus, or which were made possible by the separation of isotopes or by the production of artificial elements. The inclusion of an article on Low Temperature Phenomena is justified by the fact that some of the more important experiments described involved the use of separated isotopes. We can best give an impression of the quality of these articles on physics by stating that, although the standard naturally varies, the best of them are similar to those found in the *Reviews of Modern Physics*, and that several are by authors who have contributed to that journal.

The year 1950 was one of great activity in physical research, and it would be difficult to find space even for a list of the important experiments described in this volume. The most significant work, probably, was that done with mesons artificially produced in synchrocyclotrons and synchrotrons. The existence and some of the properties of these particles had been discovered by cosmic-ray physicists, but the larger numbers of particles available in the laboratory, as well as the possibility of producing them by simple reactions, and by more than one reaction, led to a much more complete understanding of their properties. In particular the existence of the π^0-meson was definitely established, and it became possible to assign spins and parities to the π-mesons. In other fields we may mention that considerable progress was made in the development of the shell theory of the nucleus, that fundamentally important precision measurements were made of the magnetic moment of the proton, that several new methods for measuring the masses of isotopes were put into operation, and that Lamb and Retherford published detailed accounts of their work on the hydrogen spectrum, which had such important repercussions on fundamental theory.

When we come to consider the contributions outside the field of physics, the reason for the inclusion of The Chemistry of the Actinide Elements (from actinium to californium) is easy to understand. Unfortunately, this particular contribution approximates very closely to a mere list of references, since one hundred and fifty papers are dealt with in about thirteen pages and the majority are summarized in one or two sentences. An article on Isotopic Tracers in Chemical Systems, after mentioning the most likely sources of experimental error, deals briefly with over two hundred contributions on isotopic exchange reactions, isotope effects, organic and inorganic reaction mechanisms, and surface phenomena. Analytical Nuclear Chemistry describes work on some of the new methods of analysis which have been developed as a result of recent advances in pure and applied nuclear physics, including isotope dilution analysis, and activation analysis.

Progress in Metallurgy deals with the effects of irradiation on metals, with the properties of metals important in atomic energy, including uranium and thorium, with the application of radioisotopes to metallurgical investigations and, briefly, with the properties of metals which may be used in the liquid state for cooling nuclear reactors. Two short articles review The Electromagnetic Separation of Stable Isotopes (and some of their applications) and The Chemical Separation of Stable Isotopes.

The applications of isotopic tracers to biological, medical and agricultural investigations are dealt with in three articles, in which, all together, 596 papers are summarized in 70 pages of text. These articles give a general impression of being extremely authoritative, but they are so condensed that they are almost completely unintelligible to a specialist in a physical science. The same is true of two articles on the Biological Action and the Genetic Effects of Radiation.

The authors of the separate articles were asked by the editorial committee to cover not only the chief developments during 1950, but also " sufficient background material to make possible a critical appraisal of the current status of the subject matter covered ". Most of the authors have had indifferent success with the second part of this request, and the majority of the articles might be described as progress reports for 1950. Such reports would have been extremely useful in the early months of 1951, but the production of a volume of this size cannot be done so quickly, and the value of much of the book has diminished before publication. It is only fair to say that the contributors to the first volume have had a particularly difficult task, and that, as the *Annual Review* continues to be published, it will become much easier for a writer to fill in the gap since the preceding article on his particular topic.

The policy of covering so wide a field in one volume has introduced its own difficulties. It is natural to welcome anything contrary to the general tendency to specialization. The

development of atomic energy has had an effect opposite to this trend, for it has forced many specialists in engineering, chemistry, biology and other sciences to learn a considerable amount of nuclear physics. Similarly many physicists who have had to collaborate with workers in other fields have had to take a broader view than they would otherwise have done. The articles in this book are, however, written by specialists for specialists, and it is natural to wonder whether they should not have been published in the existing reviews. No one could doubt the value of an annual publication in which the various subjects which make up 'nuclear science' were discussed in an authoritative but intelligible manner. If, however, it were the object of the editors to provide such a volume, more attention would have to be given to the art of exposition, the ground covered would have to be limited, and the style would have to approximate more closely to that of a textbook. We do not know whether this is the intention of the editors; certainly it would be impossible to have authoritative, complete, and concise review articles, such as the best in this volume, and at the same time make them all readily understood by any one reader. Perhaps the disadvantages of attempting so much have been appreciated by the editors, for we notice from the contents list of next year's volume that a smaller number of articles will be published, and that they will be almost exclusively confined to physical subjects.

This is hardly a book which many would wish to buy, but should be extremely valuable in a library. The book is well produced and its price is reasonable, considering its size.

W. J. WHITEHOUSE.

Mixtures: The Theory of the Equilibrium Properties of some Simple Classes of Mixtures, Solutions and Alloys, by E. A. GUGGENHEIM. Pp. x+270. (Oxford: University Press, 1952.) 42*s*.

This book contains a readable but extensive survey of a wide field. Electrolytes, adsorption phenomena and colloids are omitted altogether, and the solubility of non-electrolytes (dealt with recently by Hildebrand) is only touched upon. The wide variety of topics treated is evident from the chapter headings: Chapters I and II assemble and outline the derivation of various general formulae from thermodynamics and statistical mechanics. Chapters III to V deal with solutions and introduce the general 'quasi-chemical' method of dealing with mixtures used throughout, its relationships with the other available methods (combinatory, Bethe, Kirkwood) being clearly and concisely explained in Chapter IV. Chapter VI deals with lattice imperfections, but, in its present form, is so sketchy as to be worthless; the formulae derived seem only to say that the probability of a given type of imperfection is proportional to the Boltzmann factor associated with the energy required to create it! Chapter VII deals with alloys forming superlattices, Chapter VIII with gaseous mixtures, Chapter IX with surfaces of liquid mixtures, and Chapters X to XII with various cases in which the components of the mixture are of different shapes and sizes. This last field is still in an early stage of development, but the results obtained so far are encouraging.

Among the defects that might be attended to in a later edition I should like to mention the following: (*a*) There seems to be much unnecessary repetition, sometimes almost word for word, of material in Chapters IV and VII. Compare, for example, sections 7.15 and 4.23 or 7.17 and 4.25. (*b*) In section 4.28 the author begins by considering certain old measurements, then rejects all modern work except that of Scatchard *et al.*, but then finds various reasons why the mixtures used by Scatchard are unsuitable as checks of the theory! A fuller explanation of the rejection of other modern work seems called for. (*c*) I see no logical reason for putting the last three sections of Chapter VII in an Appendix. (*d*) Some confusion of thought is evident in the discussion of the probable reliability of the various types of approximation. This is a topic of enough importance to merit a chapter to itself; instead, the discussion is scattered *passim* through the text. I have two major criticisms of this discussion: (i) the author does not seem to distinguish clearly between quantities that are likely to be reproduced well by a fairly crude treatment (such as the free energy of mixing) and those for which nearly all approximations are unreliable (such as the location of the Curie temperature and the form of the specific heat anomaly); (ii) the author comes out strongly against making deductions from expansions of the partition function in powers of w/kT, w being the interaction energy, and correctly shows,

in a number of cases, that, although the successive approximations obtained by stopping at various terms may agree well with one another, they nevertheless do not converge well to the final result. He omits to mention that precisely the same criticism can be made of methods that consider progressively larger groups of sites, such as Bethe's first and second approximations and Guggenheim and McGlashan work on extensions of the quasi-chemical method to groups of three and four sites. It is vitally necessary to distinguish between cases in which the agreement of two or three successive approximations indicates that one really is approaching the correct consequence of the model and cases in which no such deduction is possible.

These, however, are defects that would not be difficult to put right and do not alter the fact that the book contains some extremely interesting material. Of particular interest is the account in Chapter VII of the application of the quasi-chemical method, extended to a group of four sites, to a discussion of the three alloys of copper and gold. The lattice is face-centred, so four sub-lattices, not two, are involved, and earlier theories were unable to give a consistent picture of all three alloys ($AuCu_3$, AuCu, Au_3Cu) within one framework. The account of solutions of high polymers is well written and interesting.

The book is likely to become a valuable work of reference. The printing and binding are, as usual in this series, of a high standard and there are remarkably few misprints. The index is adequate. H. N. V. TEMPERLEY.

The Physical Principles of Thermodynamics, by R. A. SMITH. Pp. xiii + 280. (London: Chapman and Hall, 1952.) 30*s*.

This is an agreeably written account of the fundamentals of classical thermodynamics. There are quite a number of books that could be described in these words; they are divided on the one hand into those for chemists and those for engineers, and on the other into elementary and advanced. The title page of this one says it is for students of theoretical and experimental physics, and one is glad to be able to endorse this. It leans on the whole towards the topics suitable for the 'chemistry' compartment of the above classification, but is really written with the outlook of the physicist. Its standard would be that of the student working for his degree.

The book deals with the usual thermodynamic functions and their obvious applications, devoting a chapter of about 20 pages to low temperature; here the Linde and Claude types of liquefier are sketched, higher order phase changes are discussed, the demagnetization technique of cooling is described, and the measurement of temperatures below 1° K considered.

Within its range the book is careful, accurate and easy to read. The author has chosen, however, to eschew all mention of the statistical mechanical approach, and this occasionally makes the treament both obscure and incomplete, particularly when dealing with the laws of radiation. Possibly he envisages a companion volume to deal with this aspect. If so, we shall welcome it. J. H. A.

The Theory of Relativity, by C. MØLLER. Pp. xii + 386. (Oxford: University Press, 1952.) 35*s*.

Professor Møller writes that this "is primarily a textbook for students of physics whose mathematical and physical training does not go beyond the methods of non-relativistic mechanics and electrodynamics". He is presumably following the continental usage in which 'physics' is taken to include what we in this country usually distinguish explicitly as mathematical physics or, still quite commonly, as applied mathematics. Students of other kinds of physics would scarcely have time or inclination to read a book on this subject of nearly 400 pages containing quite a lot of rather heavy mathematics. In any case, their requirements in this subject are for a grasp of its basic principles and aims and then for certain fairly simple formulae in special relativity which they do have to apply in various parts of their work. These, in the reviewer's expectation, they will not readily find in the present book. All that they need does seem to be there, but it is not easy to pick out and the author does not give them much guidance in looking for it.

As for the student of mathematical physics, it is very difficult to assess what he would get by working unaided through this book. Professor Møller states that it is "an extended

version of a course of lectures " which he has given during the last twenty years. One must conclude that he has found this presentation to have successful results. But one considers that such results would still require some lectures, as well as the reading of the book, if only to supply the right distribution of emphasis which does seem to be lacking in the printed version.

A good presentation of a subject like special relativity or general relativity ought to give it a certain *form* as a whole. There are admittedly several possible forms to choose from depending on the point of view and mathematical treatment which may be adopted. But one believes that the student ought to acquire an appreciation of, and a feeling for, one such form, before he is asked to think very much about either the foundations of the subject or about other possible forms. Otherwise he probably never will see the subject as a whole nor be convinced that it is worth while thinking about these other matters. At any rate in the judgment of the present reviewer, Professor Møller's treatment is somewhat lacking in this essential regard. If so, it is probably only because he tends to be overconscientious about the trees and so does not sufficiently often re-call attention to the existence of the wood, particularly those tracts of it that lie outside his present exploration.

On the other hand, any reader who has already got some adequate general conception of relativity theory in one of its existing standard presentations will find here much valuable material of all kinds. Some of it will stimulate his thoughts about the foundations. This is particularly true of Chapter VIII on the foundations of the general theory, with its treatment of what is really Mach's principle (though, so far as I have noticed, Professor Møller does not say this and does not mention Mach in either of his two indexes), and with its interesting treatment of a rotating frame of reference (though the notion of ' standard measuring rods ' and ' standard clocks ' seems to need more examination). Some of it will provide him with alternative formulations of important sections of the subject ; here, particular attention may be drawn to Professor Møller's stress upon the physical difference between space and time and his frequent use of vectors and tensors in 3-space instead of 4-space. Some of it will acquaint him with useful generalizations of particular results as, for instance, the most general Lorentz transformation given in § 13. Also Professor Møller includes a number of important results that have not found their way into most other textbooks on the subject, such as certain variational principles and such as the ' Thomas precession ' treated in § 22 (though Thomas is among the several authors whose initials have been arbitrarily changed for the purposes of this book !).

In a work intended for physicists, it is strange to find that the physical interpretation of some of the results is not given more attention. For instance, there is a well-known interpretation of the gravitational red-shift which does not seem to be mentioned. Also the description of the situation as regards the three ' crucial tests ' of general relativity is not up-to-date. What interests relativists is that the one test, concerning the advance of the perihelion, that does not depend upon radiation-properties seems to give satisfactory results (particularly in the work of G. M. Clemence, not mentioned in the book), whereas the other two that do depend upon radiation-properties are still in doubt. Contrary to what is said on p. 348, the " agreement with the observations " in the case of the Einstein red-shift is altogether uncertain.

In spite of the criticisms to which it is open, the book is a notable addition to the literature of relativity theory. Anyone concerned with any part of the subject to which Professor Møller has devoted particular attention will certainly find that he has something highly illuminating to say about it.

W. H. McCREA.

Valence, by C. A. Coulson. Pp. x + 338. (Oxford : University Press, 1952.) 25*s*.

The publication of Professor Coulson's new book is something of a scientific event. In spite of the great strides which theoretical chemistry has made since the advent of wave mechanics there has been no book in the English language to which the student could refer for a clear and authoritative introduction to the modern valency theory. In a theoretical discipline largely based on mathematical approximations a little learning is more than usually dangerous for two reasons : first, it tends to divide chemists into warring camps, each supporting those theoretical ideas with which they are more familiar and suspecting or openly condemning methods which they do not really understand; secondly

the concepts of theoretical chemistry can only bear fruit in new discovery if they are used with critical discrimination based on a careful study of the different methods of approach and the relation between them. As Professor Coulson says in his preface, the subject matter of valency has been, and still is, changing rapidly as befits a living discipline, and the tremendous growth of the subject has led pessimists to the view that valency theory is too large a subject to be compassed at the student level in a single volume. However, if the student cannot now be led across the gulf between empirical chemical theory and an understanding of quantum chemistry, there is little ground for hoping that he ever can.

Physical chemists, therefore, have every reason to be grateful to one of the world's leading authorities, who is also a master of clear exposition, for accepting this challenge. Professor Coulson's book succeeds admirably in its intention, which is to give the student a balanced and intelligible account of the modern theory of molecular structure. The author's aim is to show how the two principal techniques of quantum chemistry, the molecular-orbital method and the valence-bond method, have been used to account for the principal rules of valency, and to solve problems which were beyond the scope of the older electronic theory. The necessary wave-mechanical theory is developed with admirable clarity right from the beginning, and is applied not only to diatomic and polyatomic molecules, but to aromatic systems and metals as well. And the treatment is by no means a bare outline. Where another author would have yielded to the temptation to say " It can be shown by quantum-mechanical arguments that . . . ", Professor Coulson gives the details, sometimes in small print, but keeping nothing except the bare arithmetic up his sleeve. Another refreshing feature of the book is its complete freedom from partisan opinion. As the author explains, both the two main methods of quantum chemistry are now sufficiently well understood for us to recognize the folly of trusting to either alone. Having made important original contributions to both valence-bond and molecular-orbital theory, Professor Coulson is in an almost unique position to make a fair comparison of the two approximations.

In a book of three hundred and thirty pages it would be unreasonable to expect anything like an exhaustive survey of structural chemistry, and a selection of material is inevitable. Most of the discussion is rather general in character, and the author has, quite rightly, attempted to avoid controversial issues in the main body of the book. (The twelfth and last chapter differs from the rest in that it deals with topics such as hyperconjugation and hindered rotation where current opinion is by no means unanimous, and where further progress is to be expected in the future). The compounds which are chosen for illustration are chiefly those of the light elements, notably carbon, as atoms of small atomic number are in some ways easier to understand than heavier atoms, and consequently more theoretical work has been done on them. One does feel, however, that a little more space might have been found for drawing attention to some of the less well understood structural facts of inorganic chemistry. The complexes of the transition elements, for example, show a number of puzzling features whose understanding may well require some new theoretical ideas. We may hope that these will be forthcoming when the next edition of the book appears.

The presentation is admirable and reveals the hand of an experienced teacher. The arguments are well signposted, and the mathematics is never allowed to obscure the physical ideas. There are plenty of diagrams to help the more geometrically-minded, and enough tables to remind the reader of the magnitudes which are under discussion. The book will be indispensable to all chemists who are interested in the foundations of their subject, and to all physicists who wish to learn about the application of physics to chemistry.

H. C. LONGUET-HIGGINS.

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ABSTRACTS FOR SECTION B

On the Design of a 15 MeV β-Ray Spectrometer, by R. L. Graham and O. Klemperer.

ABSTRACT. The design of a lens spectrometer with respect to economy in power consumption and copper weight, resolving and collecting power has been studied by small scale experiments. The construction and performance of a large β-ray spectrometer is described which focuses 15 MeV electrons when its coil is energized by 50 amp at 360 volts. The coil is cooled by a flow of 12 gallons of water per minute. Maximum resolution and collecting power of the spectrometer are 0·8% and 0·6% respectively. The influences of spherical aberration, source and window size on the resolution of the thick lens spectrometer are discussed.

The Wave Aberration Associated with Skew Rays, by H. H. Hopkins.

ABSTRACT. It is shown that invariant foci exist for skew rays, and simple formulae are obtained for the change in aberration relative to those foci due to refraction or reflection at an interface. The formulae are valid for any form of interface. Expressions for focal shifts are obtained together with check formulae.

The method makes possible the analysis of the aberrations along skew rays as they arise at individual surfaces.

Ultrasonic Absorption in Mixtures of Ethyl Alcohol and Water, by L. R. O. Storey.

ABSTRACT. The pulse method has been used to measure the absorption of ultrasonic waves in mixtures of ethyl alcohol and water at the three frequencies 22·5, 37·5 and 52·5 Mc/s and at the temperatures 0°, 10° and 25°C. The results confirm the findings of earlier workers that in both the pure liquid and the mixture there is more absorption than can be explained by the classical mechanisms of viscosity and heat conduction. When the composition of the mixture is varied a maximum of absorption is found for a mixture containing 20 to 30 molar per cent alcohol. The magnitude of the absorption at the maximum increases rapidly as the temperature is lowered. Over a range of compositions in the neighbourhood of the maximum the value of α/f^2 (α is the absorption coefficient and f the frequency) is found to decrease as f increases.

It is known that the extra absorption in the pure liquids can be explained in terms of a compressibility with a finite relaxation time, but this mechanism alone cannot account for the behaviour of the mixture. It is suggested that here the additional absorption is due to interaction between the molecules of the two components, leading to the partial development of a structure or complex in the liquid with a composition of four molecules of water to one of alcohol. The thermal properties of the mixture indicate that such interaction occurs, and the suggestion for the composition of the complex is supported by the fact that the freezing-point curve for the mixture shows that a structure with this composition is formed in the solid phase.

The extra absorption found by other workers in several similar mixtures can be explained, at least qualitatively, in the same way.

Crystal Structure and Antiferromagnetism in Haematite, by B. T. M. Willis and H. P. Rooksby.

ABSTRACT. The crystal structure of haematite has been investigated by x-ray analysis in the temperature range 20°C to 950°C. On cooling through a temperature of approximately 675°C the rhombohedral unit cell undergoes a sudden expansion along the triad axis direction. The results are interpreted in terms of an antiferromagnetic behaviour of pure haematite, with a Curie point at approximately 675°C. It is shown that the behaviour is analogous to that of the cubic oxides FeO and Fe_3O_4, which exhibit structure cell deformations on cooling through certain critical temperatures.

The Thermal Oxidation of Tin, by R. K. Hart.

ABSTRACT. Electron diffraction experiments show that when tin foil is thermally oxidized in air an amorphous oxide layer is formed at temperatures up to 130°C. Above 130°C only crystalline SnO and SnO_2 are formed.

Accommodation Kinking Associated with the Twinning of Zinc, by A. J. W. Moore.

ABSTRACT. Small changes of slope on cleavage surfaces of zinc single crystals have been measured by a stylus type profile recorder (Talysurf) and compared with photomicrographs of the same areas. Lattice bending through angles up to approximately 0° 47′ which relieves the localized shearing stresses due to twinning has been observed.

Other examples show firstly one twin with two associated accommodation kinks, and secondly a twin and its associated kink band occurring within the kink band associated with a second parallel twin.

Transistor Action in Lead Telluride, by C. A. Hogarth.

ABSTRACT. Transistor action has been observed in single crystals of lead telluride, examined at 90° K. Properties of the units are described and it is found that values of current gain greater than unity may be observed.

Electrode Phenomena in Transient Arcs, by J. M. Somerville, W. R. Blevin and N. H. Fletcher.

ABSTRACT. High current, short duration arcs between various metals in air have been studied by means of a Kerr Cell camera and by observation of the marks left on the electrodes. Growth of these marks with arc duration is discussed and shown to be due to heat conduction in the case of the anode marks and to motion of the emitting areas in the case of cathode marks. Cathode spot temperatures are frequently too low for thermionic emission even on metals of high boiling point. At very high rates of current rise both anode and cathode marks are multiple.

The Measurement of the Angular Diameter of Radio Stars, by F. G. Smith.

ABSTRACT. A new method of using interferometers to measure the diameters of radio stars is described. The method is based on the use of the phase-switching system, and has several practical advantages over previous methods.

Measurements which have been made on the radio star (23.01) in Cassiopeia, indicate that the radio star has an angular diameter of about 5′ arc, the exact value depending on the distribution of brightness across the star. A gaseous nebula reported by Baade, in the same position as the radio star, has about the same diameter. Some measurements have also been made on the radio star (19.01) in Cygnus, and this star appears to have an angular diameter of about $3\frac{1}{2}$′ arc.

Tropospheric Wave Propagation in a Duct of Non-Uniform Height, by H. G. Hay and R. S. Unwin.

ABSTRACT. This paper describes an approximate method for extending the simple mode theory of tropospheric refraction (Booker and Walkinshaw 1947) to cover situations where the refractive index profile varies with range. The transmission path is divided into a number of zones, in each of which the variation in the refractive index profile is small. In each zone the profile is then taken to be independent of range, and given by the mean profile throughout the zone. The duct-width d and the lapse-rate of modified refractive index at infinite height κ_∞ are found for each zone and are used, together with published propagation curves, to evaluate the distribution of radio field strength in the region beyond the horizon of a transmitter. Although the method is a general one, the final formula refers specifically to situations in which the field can be adequately described by the first mode alone, and where the mean profile of refractive index in each zone may be represented analytically by a one-half power law curve.

An example of the application of the method to a practical situation is given. This shows good agreement between experiment and theory, and demonstrates the significant effect of changes in the refractive index profile on the level of field strength. The limitations of the method are pointed out, and its extension to cover other types of refractive index profile and higher order modes is outlined.

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